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VACUUM
PHYSICS

A symposium held by the
Midland Branch of The Institute of Physics in Birmingham
on 27 and 28 June 1950

JOURNAL OF
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INSTRUMENTS

SUPPLEMENT NO. 1

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Editorial Note

Iτ is the practice of some of the geographical branches and subject groups of the Institute of Physics at home and overseas to hold occasional two- or three-day meetings on some particular branch of applied physics, with the object of bringing together those working on the subject in industrial, government and university laboratories. Some of these symposia have been published by the Institute in its *Physics in Industry* series—for example, *The Measurement of Stress and Strain in Solids*, 1948, and *The Acceleration of Particles to High Energies*, 1950. It has now been decided by the Board of the Institute of Physics that in future symposia of this nature would more appropriately appear as supplements to one or other of its journals—*Journal of Scientific Instruments* and *British Journal of Applied Physics*. In accordance with this decision, the first supplement to the latter journal was published in March and contained the papers contributed to the symposium on the Physics of Lubrication, held in July 1950 and the discussion on them.

This, the first supplement to the *Journal of Scientific Instruments*, contains the papers presented at a symposium arranged by the Midland Branch of the Institute of Physics and held, at the invitation of Professor M. L. Oliphant, in the University of Birmingham on 27th and 28th June, 1950. The discussion printed here is substantially that which took place after the presentation of the papers at the meeting but it has been edited, and where necessary revised, by the speakers themselves.

I wish to record my grateful thanks to the referee, who prefers to remain anonymous, for his expert advice, to the authors for revising and often reducing their scripts, to Mr. L. Riddiford for assistance in editing the discussion, and to Unwin Brothers Ltd., The Gresham Press, for their co-operation and support at every stage of the production of this supplement.

The result, it is hoped, is a clear and concisely written progress report on vacuum physics and its applications in other branches of science and in industry. These applications are growing rapidly and vacuum techniques are now used in a wide range of industries and laboratories. Those responsible for this apparatus and plant will find up-to-date information not only on the means of creating and maintaining vacua, but also on the equally important ancillary equipment for measuring those low pressures and for detecting leaks. They will also find an account of some interesting and important industrial applications of high vacuum techniques. The committee which issued the invitations to read papers could not attempt to cover all aspects of this subject in two days, but the papers presented covered a sufficiently large cross-section to be helpful to all those concerned with vacuum physics or who wish to keep themselves informed of the recent developments.

<div align="right">H. R. LANG</div>

Institute of Physics, London
July 1951

CONTENTS

Modern vacuum pumps

By D. R. GODDARD, M.Sc., A.Inst.P., British American Research Ltd., Wishaw, Lanarkshire

The two distinct classes of pump, the mechanical and the vapour pump, are discussed in a general rather than a detailed theoretical manner with a view to examining the factors influencing performance and the more important design features.

The different types of rotary pump are compared and their performance examined in relation to the requirements of capacity, ultimate pressure and the pumping of condensable vapours. The mechanism of vapour pumps is discussed in relation to the design of ejectors and diffusion-condensation pumps. A comparison of oil and mercury pumps is made and the requirements of capacity, ultimate pressure and backing pressure characteristics discussed.

The rate of development of the new industry of vacuum engineering has been determined by the evolution of the different types of vacuum pump and their applications. This has been largely a consequence of the demands of electronic and chemical industries and there are now few fields in which vacuum has not found some application.

By "vacuum" we refer to any reduced gas pressures with respect to normal atmospheric pressure, but it is in the higher vacuum range, that is, in the use of pressures below 1 mm of mercury that the most significant developments have been made. The history of the vacuum pump dates from Torricelli's studies of the barometer and Guericke's piston pump of the seventeenth century, but the modern rotary and vapour pump arise principally from the work of Gaede and Langmuir.[1] It is perhaps remarkable that there has been so little change in the fundamental principles of pump design from those involved in the original Gaede rotary pump or the Langmuir condensation pump. The impetus of developments over the last twenty years, and especially the war years, has produced larger and larger pumps of designs which are continually being improved, and there are now available commercially very comprehensive ranges of pumps for any known application. It is these modern pumps, their design features, and the factors influencing their performance that is the subject of this review.

There are two distinct classes of pumps. Mechanical and vapour pumps. Mechanical pumps are of the rotary or of the molecular drag type. Reciprocating pumps are rarely used. Vapour pumps utilize condensable vapour as their driving force, and are divided into the ejectors and the vapour stream pumps. The latter are better known as the diffusion-condensation, or simply diffusion pumps.

In the range 760 mm to 0·1 or 0·01 mm of mercury conditions of viscous flow exist, i.e. the mean free path of the gas molecules is small so that collisions between themselves are more frequent than collisions with the walls of the vessel containing them. Below 10^{-3} mm the reverse is the case. Mechanical pumps and ejectors are generally designed to have their greatest efficiency in the viscous flow region and diffusion pumps in the molecular flow region, and, although mechanical pumps and ejectors can be made to work at pressures well into

the molecular region and diffusion pumps up into the viscous flow region, it is general practice to use a combination of pumps of both classes for pressures below 10^{-3} mm.
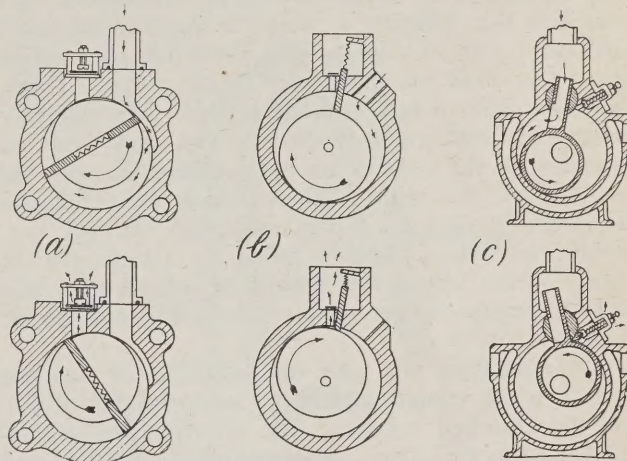

Fig. 1. Types of rotary pumps: (a) rotary vane-type; (b) fixed spring-loaded vane type; compression at each revolution; (c) Kinney type; the free cylinder follows the motion of the rotor and maintains a seal on the wall of the housing; the vane moves in the manner of a piston rod on a crankshaft

ROTARY PUMPS

The normal rotary pump consists of a rotor which rotates in a housing or stator of larger diameter than itself so that a point on its circumference is always in contact with the housing to make a seal. A sliding vane cuts off a portion of the crescent-shaped volume swept out and the gas therein is compressed and forced out of a non-return exhaust valve to the atmosphere. Further air is drawn in by the expansion on the other side of the vane ready for compression and ejection on the next cycle. The pump is either immersed in or continuously fed with oil for sealing, lubrication and cooling. Clearances between moving parts are kept to a few thousandths of an inch by accurate construction to ensure a good seal. The types of rotary pumps are shown in Fig. 1.

With each type of pump there is a constant volumetric displacement per cycle and different designs vary only

in capacity or rate of pumping and the volumetric efficiency obtained at various pressures. The ultimate pressure and efficiency are limited by (a) the practical limits of accuracy in machining and subsequent wear which allow some leakage of gas back past the vanes and rotors into the system; and (b) the vapour pressure of the oil and, what is more important, contamination with condensable vapours.

By compounding two or more pumps in series the effects of the first limitation may be reduced so that pressures of less than a micron may be obtained. Single stage pumps will produce pressures of 5 to 10 μ on the average. The several stages may be incorporated into one pump body with a common driving shaft. Even with compound pumps, however, small quantities of water vapour will raise the ultimate pressure considerably and contamination is the most serious limitation. To minimize this difficulty a moisture trap can be used, such as a trap of phosphorous pentoxide over which the air is dried on its passage to the pump. Traps, however, have to be constantly refilled. The difficulty with water and other vapours is that they form an emulsion in the oil and are never eliminated with the air. There are rotary pumps which will eliminate condensable vapours; for example, the German Leybold's gas ballast pump in which air is bled into the pump on the exhaust side during the compression cycle so that the expelled gas-vapour mixture does not become saturated.[2]

Oil separators are used for attaching to standard pumps which contain electric or steam heaters to boil off the absorbed vapours. The oil is cooled and recirculated to the pump. Alternatively a centrifuge may be used to separate the oil from the condensed vapours. These pumps are widely used in the vacuum drying and impregnation industries. Some rotary pumps have been run with hot oil at about 110° C so that the water vapour passes right through. This method can be used to handle several pounds of water vapour per hour whilst maintaining a free air pressure as low as 30 μ. The oils normally used for rotary pumps are good quality light petroleum oils with the higher vapour fraction removed. For rough vacuum work practically any oil can be used.

Rotary pumps, e.g. Geryk type, are available which will produce pressures down to 10^{-6} mm of mercury as measured on a McLeod gauge. The average rotary pump, however, will give 1 to 10 μ which is well within the range of backing pressure required for most diffusion pumps, and, unless the rotary pump is used alone, lower pressures are not required. Rotary pumps are made in a wide range of sizes with volumetric displacements from a fraction of 1 ft^3/min up to about 700 ft^3/min. Many of the larger pumps have two or more stages in parallel to increase the capacity and there are also series parallel arrangements for high capacity with lower ultimate pressures.

The choice of a rotary pump depends on the particular application. The factors to be considered are: (a) working pressure range required; (b) capacity required—dependent on size of vessel to be pumped, time to pump down, or size of vapour pump to be backed; (c) whether vapours are to be pumped or not; (d) minor details such as physical size and amount of noise or vibration produced may be important.

In addition to the rotary pump there is the lesser-known molecular drag pump which was also introduced by Gaede. The principle of its operation is the dragging effect exerted on gas molecules by a rapidly-moving surface close to a stationary surface. By the use of high speed rotors or disks (about 10 000 r.p.m.) in stators with highly-finished surfaces and small clearances, speeds of up to 80 l/sec and very low ultimate pressures (less than 10^{-6} mm of mercury) have been attained.

These pumps have the advantage that they will pump gases and vapours so that traps are not required, and they pump heavier gases faster than light (opposite to diffusion pumps) which has suggested their use on cyclotrons where it is necessary to maintain a certain pressure of hydrogen or deuterium in the presence of a minimum of heavier residual vapours. The precision required in construction has, however, discouraged their use except for laboratory purposes in favour of the simpler rotary pumps.

VAPOUR PUMPS

In all vapour pumps such as steam and oil ejectors, mercury and oil diffusion pumps, a stream of high-velocity vapour is the driving force. In order to review the design features and performance of vapour pumps it is necessary to consider the action of the vapour stream. Briefly at this stage a steam and oil ejector can be regarded as a nozzle and exhaust tube attached to a vessel under evacuation (Fig. 2) and a diffusion pump as the same, with the addition of a condensing surface to condense the vapour in the exhaust tube (Fig. 3). A source of vapour is obtained from a boiler which in the case of the oil pump is integral with the pump and in the case of the steam ejector is usually obtained from an ordinary steam supply. There has been much controversy in the past as to the mechanism of vapour pumps—whether or not the diffusion pump of Gaede works on the same principle as the condensation pump of Langmuir. Much of this appears to be due to the confusion of the interpretations given to the processes of diffusion and jet action and has been explained to some extent by the work of Molthan and Alexander, and others.[3]

The jet nozzle is used to transform the pressure energy of the vapour into velocity energy in a directed stream. At high gas pressures in the viscous range and vapour velocities less than sound (i.e. comparable to molecular velocities) the vapour stream is in the form of a confined jet with eddies at the gas vapour boundary which entrains the gas by suction into the stream and compresses it down the exhaust tube. A high vacuum

cannot be reached and the pressure is limited by the back scattering of vapour when the gas pressure becomes less than the vapour pressure. In actual steam ejectors and oil ejectors the vapour velocity becomes supersonic as the pressure is reduced and very little is scattered back, which allows much lower pressures to be reached.
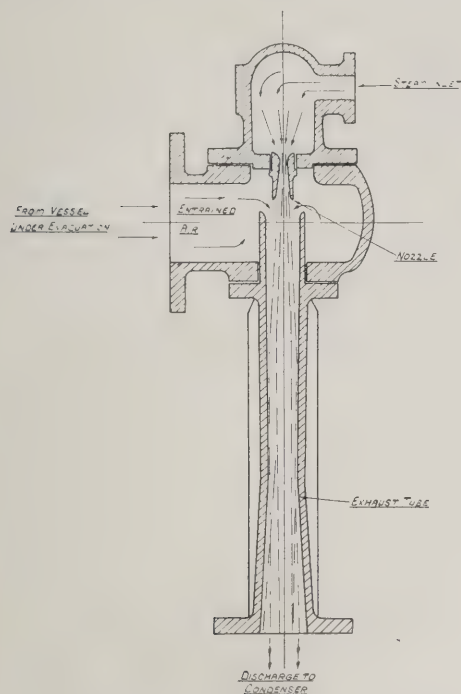

Fig. 2. Diagrammatic sketch of steam ejector

In the molecular flow region below 10^{-3} mm of mercury the vapour spreads out of the nozzle at supersonic speeds so that a small fraction is actually flowing backwards. However, Alexander[4] has shown experimentally that the bulk of the vapour flows downwards and the mean free path of the gas molecules in the outer fringe is still large so that they can diffuse through into the main stream. The action in diffusion pumps (and in ejectors at low pressures) is that the vapour stream sweeps the gas downwards by imparting to it part of its own velocity. As the vapour stream expands the gas loses its streaming velocity and its density increases to a maximum. The compressed gas is then pumped away to the atmosphere by a mechanical pump. The space in the vicinity of the nozzle becomes rarefied and more gas diffuses in from the vessel being evacuated. The advantage of the supersonic flow is lost unless the vapour can be rapidly condensed when it has done its job. In the steam ejector the expansion in the exhaust tube is sufficient to condense the steam. In the oil ejector and diffusion pump a condenser is required. This was the essential feature realized by Langmuir.

Some pumps may act as ejectors in the high-pressure range and as diffusers in the low-pressure range. This is especially so with two or three stage diffusion pumps and with oil ejectors. The higher the heat input the lower the pressure at which the ejector action continues to take place in both diffusion pumps and ejectors.

Alexander has shown that for a vapour pump to work efficiently there must be a point of minimum and maximum gas density on any line parallel to the direction of the pumping from the plane of the nozzle to the condenser wall. The jet impact must be sufficient to push the gas against a pressure gradient, and therefore at high pressures the quantity of vapour must be adequate. At lower pressures the velocity of the vapour stream must be large enough to prevent back diffusion of the compressed gas. Thus the density and velocity of the vapour stream in addition to the physical design of the nozzle, the width of the diffusion slit and the disposition of the condenser wall are important factors in the design of any vapour pump
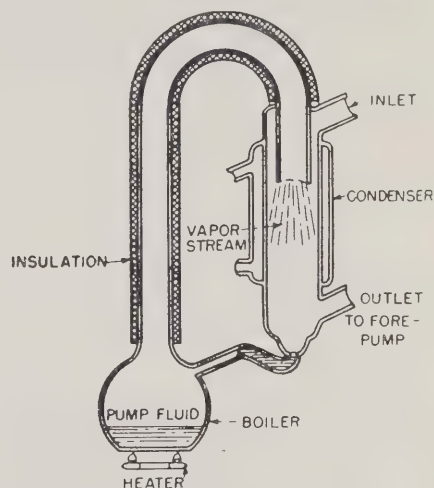

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Fig. 3. Langmuir type condensation pump illustrating principles of diffusion pump

The nozzle design determines the proportion of vapour going in a forward direction. A divergent nozzle, the longer the better, produces a stream in which the vapour molecules move in nearly parallel directions with few collisions between molecules which lead to backstreaming. The density and velocity of the vapour stream determines the pressure range over which the pump will function and the backing pressure required to deal with the compressed gas.

The principal characteristics of pump efficiency are (*a*) the ultimate pressure produced, which is limited by the vapour pressure of the pump fluid and to a certain extent by back diffusion of gas molecules as well as vapour molecules, and (*b*) the speed factor or Ho coefficient which is the ratio of the observed pump speed measured as the volume of gas pumped per unit time at a given pressure to the theoretical speed of diffusion through an orifice with the same area as the pump slit or throat

(11·7 l/sec/cm² for air). The speed factor is generally below 0·5 due to the impedance of the pump head and the back streaming of both gas and vapour. The most significant factor influencing the performance of a vapour pump is the heat input. It is the heat input which determines the velocity and density or the energy of the vapour stream, and which consequently determines the critical backing pressure and pumping speed. The efficiency or Ho coefficient is influenced by heat input
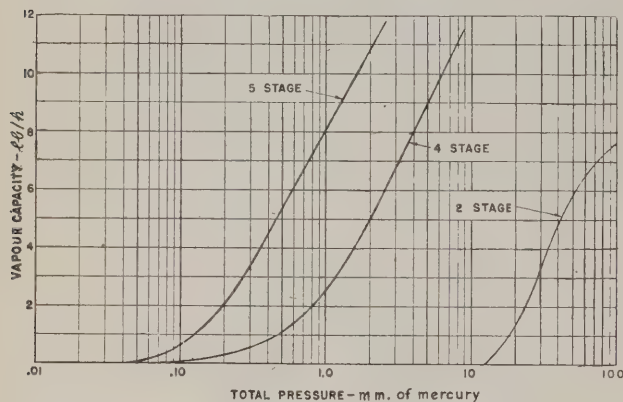
Fig. 4. Speed versus pressure data for three representative commercial steam ejectors

and, as fine side pressures are affected by the backing pressure heat input indirectly influences the ultimate pressure.

Types of Vapour Pumps.—The steam ejector is rather outside the scope of this short review as it is used in the higher pressure ranges. However, by the use of several stages, that is, several nozzles in series with intermediate condensers, it is possible to achieve pressures of 50 μ with high capacities (Fig. 4). Many applications of these pumps have been made in the chemical industry for fractional distillation and for drying on a large scale and in the food industries. Economically steam ejectors are most efficient above $\frac{1}{2}$ mm of mercury. Below this pressure the rotary pump comes into its own. The steam ejector, of course, deals directly with condensable vapours.

The mercury diffusion pump is widely used for laboratory work and the manufacture of lamps and electronic valves. In its most conventional Langmuir form it has umbrella type annular jets and is made in either glass or steel. Other types are based on the original Langmuir condensation pump with a single jet of the divergent nozzle type pointing downwards (Fig. 3), upwards or horizontally with a surrounding condenser. The condenser surface is sometimes conical and usually water cooled. Air cooling is sometimes sufficient and may even be an advantage in some designs to ensure a high concentration of vapour at the wall to prevent back diffusion of the gas. In the umbrella type the boiler tube supplying the jet is centrally placed within

the pump [Fig. 5(a)]. In other types the boiler tube is external to the pump volume. The use of two or more stages of jets in series allows the pump to work against higher backing pressures. The number of different designs is very great as it has been general practice for laboratories to build their own pumps with their own special features. There are, however, standard ranges of mercury pumps available commercially. Most mercury pumps have a speed of only a few litres per second. There are exceptional cases where speeds of several hundred l/sec have been attained. Perhaps the most outstanding in recent years has been the Alexander type[4] based on his conclusions drawn from the theory of vapour pumps. This pump is two stage, in metal, with a diameter at the first jet of nearly 11 in and gives a speed of about 800 l/sec in the range 10^{-4} mm to 10^{-2} mm of mercury. This speed has been increased in later models. The main disadvantage of the mercury pump is that owing to the vapour pressure of mercury being about 10^{-3} mm at normal temperatures, pressures lower than this can only be obtained by the use of vapour traps above the pump (usually of liquid air). In addition, for many modern applications the pumping speeds of most mercury pumps are too low. Apart from these limitations mercury is ideal as a pumping fluid.
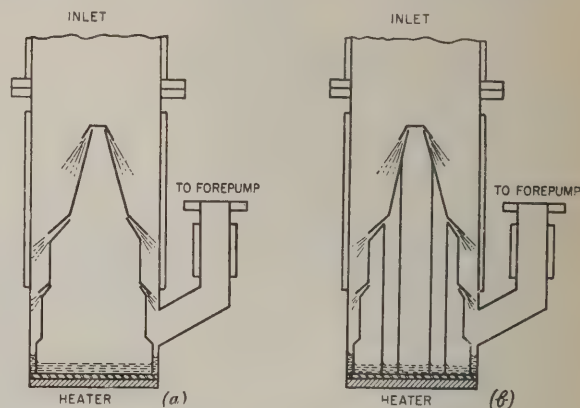
Fig. 5. Metal oil vapour pumps: (a) non-fractionating; (b) fractionating type

The advent of oils with lower vapour pressures than mercury as pump fluids due to the work of Burch in 1928–29 and later Hickmann, opened up the field of high vacuum applications considerably by the development of large oil diffusion pumps with very high capacities. The need for vapour traps was eliminated for most normal applications, but where very low pressures below 10^{-6} mm of mercury are required or when it is necessary to prevent oil vapour diffusing back into the system, a trap cannot be dispensed with entirely. Baffles of various designs are used instead of a trap to cut down the back diffusion of oil but at the expense of some loss in pumping speed.[5]

The design of vapour pumps in general is based on the same principles as apply to mercury · pumps. Dushman[6] has pointed out that, on the basis of published figures available for various types of pump construction, a given design of pump will, in general, produce a higher pumping speed with oil than with mercury. However, because of the special characteristics of the oils used the oil pump generally has its own special features of construction. The oils are susceptible to decomposition when hot and the boiler and vapour column must be large enough to give an adequate supply of vapour to the jet without the need for excessive boiler temperatures and superheating. The volume of oil in the boiler should be kept to a minimum to avoid excessive quantities of high vapour pressure decomposition products accumulating. As the latent heat of oil is low the jet system should be made of a good conducting material such as aluminium or copper so that heat may be conducted from the boiler instead of by condensation of the oil vapour to keep the jet system hot. Copper is sometimes to be avoided with certain oils as it may catalyze a cracking of the oil. It can, of course, be plated with nickel or chromium.

Oil vapour pumps made of glass are used, but most pumps of this type are made of metal—usually steel. They normally have two or three stages of jets in series and on the average require a backing pressure of 200 to 500 μ. They vary in size from 1 in diameter up to 16, 20, or even 32 in diameter with corresponding speeds from less than 10 l/sec to 15 000 l/sec.

Among the oils used the original vacuum distilled mineral oils with vapour pressures around 10^{-6}–10^{-7} mm are still the most popular. In addition to the Apiezon A and B oil there is now available a C oil which has a vapour pressure of 10^{-8} mm for use in very high vacuum work. The synthetic oils are also widely used, for example, the higher order esters such as butyl phthalate and sebacate and especially octyl phthalate and sebacate. The silicone oils are comparatively new and are claimed to have better resistance to oxidation than other oils. Chlorinated hydrocarbon oil is also very stable and does not crack easily.

The oils vary slightly according to their resistance to oxidation and to their vapour pressures. Slight differences in the properties of the oils cause small variations in speed and backing pressures required in a given pump for the same heat input. The disadvantage associated with oils in their susceptibility to decomposition may be minimized by the use of the fractionating or purifying pump [Fig. 5(b)]. The boiler or succession of boilers are so constructed that the condensed oil must first pass into the boiler section supplying the last stage of jets where the lighter components are boiled off and tend to be eliminated by the backing pump. In this way only the lowest vapour pressure fraction of oil reaches the high vacuum jet and consequently the lowest ultimate pressures can be obtained. This continuous conditioning of the oil is apparent to some

extent even in non-fractionating pumps and all oils improve with use providing they are not exposed to excessive quantities of air.

The choice of a vapour pump is governed by the following requirements: (a) speed or capacity—it is general to allow 1 l/sec speed for every litre volume to be pumped with additional speed to cope with evolved gases and to give short pumping cycles; (b) ultimate pressure—determined by the type of oil and design of pump, and (c) backing pump characteristics. The choice of rotary backing pump is very important and is determined by the critical backing pressure of the vapour pump, i.e. the pressure at which the jets break down, and by the capacity of the vapour pump. The speed of a vapour pump is usually independent of backing pressure below the critical value, but the ultimate pressure will be raised unless a good backing pressure can be maintained. Most modern vapour pumps will work against backing pressures of 0·2 to 0·5 mm, which is well within the limit of any rotary pump in a good condition.
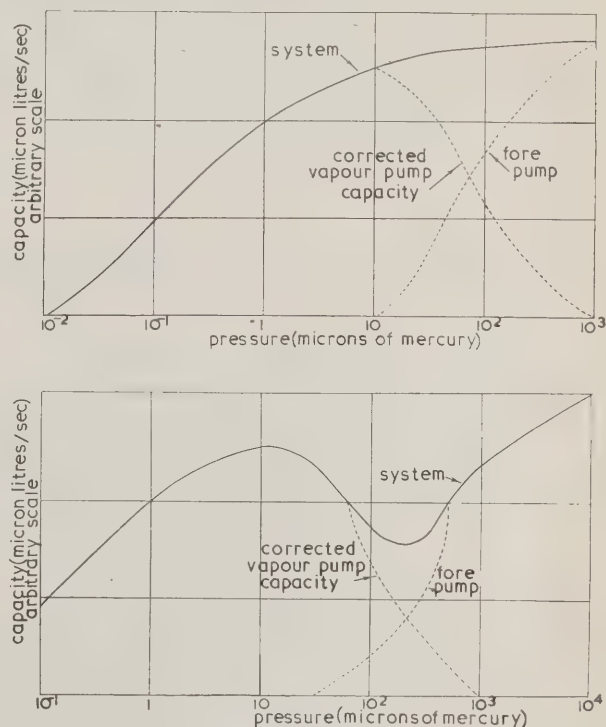

Fig. 6. Shape of capacity curves for high vacuum system (after Mellen): (*above*) with matched vapour and fore pump; (*below*) with inadequate fore pump

The capacity condition is such that in order to ensure a stable pumping system the backing pump capacity should be greater than that of the vapour pump over the critical range at which the capacity of the vapour pump starts to fall off (Fig. 6). More exactly the pumping capacity of the system as a whole, measured

at the vapour pump inlet must be increasing with increased pressure throughout the whole range from atmospheric downwards, otherwise it is difficult to achieve short evacuation times and to deal with gas bursts and pressure surges.[7] This is one reason why capacity, or, as it is sometimes called, throughput or molecular speed, is considered a better indication of pump performance than the intrinsic speed. Witty[8] has suggested that the rate of pumping should be expressed in terms of throughput or quantity of gas $P \times V$ pumped per second, which is proportional to the mass instead of in terms of speed or the volume of gas pumped per second. A point to watch in comparing pump performance curves is the method used to measure pressures. Some curves are given with McLeod gauge figures and thus only show gas pressure. Ion gauge figures show total pressures (unless a trap be used). The gas speeds are theoretically constant below the critical pressure, but the speed of gas plus vapour falls to zero at the ultimate pressure due to the equilibrium reached with the back diffusion of both gas and vapour.

The methods of measuring speed are discussed by Dayton.[9] In actual practice there may be irregularities in the speed or capacity curves due to back diffusion of gas through the vapour stream under variable conditions. Varying the distribution and intensity of water cooling, the heat input and nozzle design will help to find the cause.

The construction of the modern pumps is extremely simple, but many seem to be designed on a hit-or-miss basis, and the only justification for their existence is that they work. More investigation is needed to elucidate the mechanism of the pumping action, especially second order effects before it will be possible to predict exactly the correct design for a given performance. Experimental work such as has been used recently, namely the use of a high voltage discharge to render visible the supersonic flow conditions in the vapour stream, may make it possible to establish an empirical basis if not an exact mathematical theory of the action of vapour pumps.

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DISCUSSION

Mr. J. Grindley: It is unlikely that the curves of pump performance presented would bear a very strict comparison. There is as yet no general agreement on the precise definition of pumping speed or the conditions under which it should be measured.[1] Further, at higher pressures the form of the curve is governed by backing pump capacity, and so should be regarded as pertaining to a specific rotary and diffusion pump combination.

Has Mr. Goddard any direct evidence of the catalysis of diffusion pump oil decomposition by metals?

Author's reply: The curves presented were supplied by the manufacturers of the pumps concerned, and the inconsistencies in them serve only to emphasize Mr. Grindley's remarks. Action should be taken through an official body to standardize the nomenclature and the testing methods used to assess the performance of any piece of vacuum apparatus.

I have no experience of the catalysed cracking of pump oils, but Martin and Hill[2] cite hot copper and brass as metals which catalyse the decomposition of some oils. We have experienced trouble with aluminium jet systems and dioctyl sebacate through the formation of tarry deposits which are obviously due to thermal decomposition, but it is not known whether the process is catalysed or not.

Mr. D. Latham: Mr. Goddard follows Dushman in assigning to mercury diffusion pumps a Ho coefficient about half that of oil pumps. We have made a large number of speed measurements on both oil and mercury pumps and find that the Ho coefficients do not differ appreciably. For example, our Type 603 oil diffusion pump has a speed at 1μ of 570 l/sec, giving a Ho coefficient of 0·36, while the corresponding figures for the 6M3 mercury pump are 630 l/sec and 0·35. These pumps are both of 6 in bore and very similar general construction.

We have, however, found that the speed of a mercury pump is far more sensitive to condenser wall temperature, decreasing with increase of wall temperature. Low Ho coefficients for glass pumps are sometimes due to an appreciable temperature gradient across the condenser wall and it is possible that this effect might explain the experiments of Sutton and Pollard which Mr. Goddard mentioned. The pump demonstrated by these workers at the Physical Society's 1950 Exhibition had a Ho coefficient of 0·30 with amalgamated copper condensing walls which compares unfavourably with the figures for standard production pumps mentioned above. It should, however, be emphasized that Sutton and Pollard have been interested in the possibility of obtaining

low absolute pressures rather than high Ho coefficients. Alexander[3] quotes a maximum Ho coefficient of 0·34 for mercury pumps which he has constructed.

The ultimate vacuum of 10^{-6} mm of mercury quoted for glycerol is surprisingly low. Tests in our laboratories indicate an ultimate of about 10^{-4} mm of mercury at cooling water temperature of 20° C, which is in agreement with published values for the vapour pressure. We also found that conventional one-hit baffles did not completely suppress back-streaming, presumably owing to the high intrinsic vapour pressure resulting in an appreciable number of intermolecular collisions in the region of the baffle. A point worth mentioning in connection with the hygroscopic nature of glycerol is that the pump used for these experiments was left standing at atmospheric pressure for three weeks, after which it was found that rusting had occurred on parts of the steel jacket which had a film of glycerol left on them. The conventional fluids will, of course, prevent rusting.

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The design of industrial vacuum systems

By T. S. MILLEN, B.Sc., A.M.I.E.E., A.M.I.Mech.E., Metropolitan-Vickers Electrical Co. Ltd., Manchester

After outlining the laws governing the flow of gases at low pressures and their relation to the performance of individual components, the author proceeds to discuss a trial design. The information required is discussed and the factors which influence the final choice of components are dealt with in some detail with special reference to the basic features and the influence of operating methods and conditions on the design.
Systems of control are described and an indication is given of the methods by which automatic protection can be applied.

In dealing with any particular problem of vacuum plant application, the designer will have certain standard components available such as mechanical and diffusion pumps, valves for hand or power operation, and standard sizes of pipes and flanges. In addition to the mechanical devices certain items of electrical equipment, such as motors, heaters, contactors and switches, are needed, and these would usually be of standard types.

The present tendency is towards the inclusion of protective devices on all plants other than those of the most elementary type. This protection may be just sufficient to guard against breakdown of the equipment or may be more comprehensive to enable the plant to be operated by relatively unskilled personnel. The designer will have to determine the extent and nature of the protection needed for each plant, and as far as possible will use standard components.

The design may therefore be resolved into three main sections:—

(1) The selection of the mechanical components.
(2) The provision of electrical power in suitable forms and its control.
(3) The arrangement of a protective system adequate for the needs of the particular plant.

In the following sections the mechanical components of the system are treated in most detail and an outline is given of the form that control and protection of the system may take. Electrical components are not considered individually in order to limit the lengths of the paper.

FLOW OF GASES

The laws governing the flow of gases in the pipes, valves and other elements of the equipment are of fundamental importance. In the high vacuum field those dealing with molecular flow are of primary interest, and are based on the work carried out by Knudsen in the period 1907–1909.

It is useful to note that the frictional resistance to gas flow at normal pressures is very much less than in the region where the mean free path of the gas is of the same order as the dimensions. In general, therefore, a system design to operate efficiently under molecular flow conditions will be satisfactory when the pressures are high during the rough pumping period. The basic equation is

$$Q = \frac{P_2 - P_1}{W \sqrt{\rho_1}}$$

where Q is the volume of gas, P_1 and P_2 are the pressures at the two ends of the pipe, W is a factor based on the shape and dimension of the tube and ρ_1 is M/R_0T the density of the gas.

From this basic formula can be derived expressions for the conductance of tubes and orifices.

For a thin plate the conductance

$$F_0 = 11 \cdot 428 \, r^2 \sqrt{(T/M)} \; \text{l/sec}$$

where T is the temperature in °K, M is the molecular weight of the gas and r is the radius of the hole in the plate.
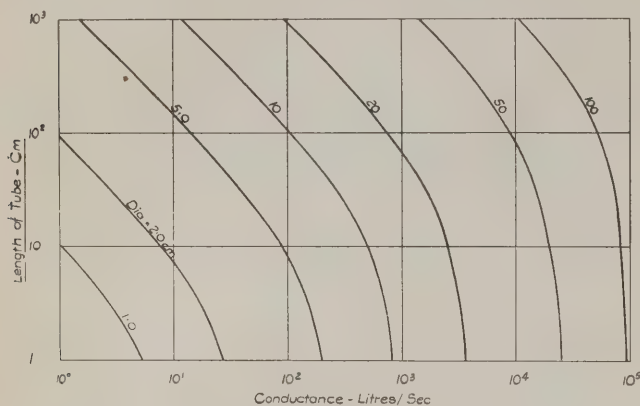

Fig. 1. Conductance U_o of cylindrical tubes at zero pressure for dry air at 20° C

Similarly for a long tube ($l/r > 100$). The conductance

$$F_t = 30 \cdot 48 \, \frac{r3}{l} \sqrt{\frac{T}{M}} \; \text{l/sec}$$

where l is the length of the tube. Other symbols as before.

From these equations an expression covering the medium length tube has been developed by Clausing, and
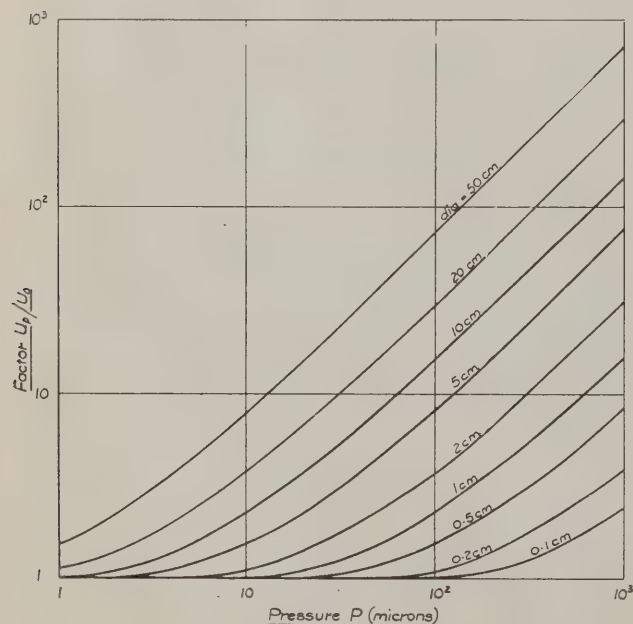
Fig. 2. Factors for converting conductance $\dot{U}_o$ at zero pressure to conductance U_p at pressure P

when plotted for air at room temperature provides the fundamental design data in a convenient form. A typical plot is reproduced in Fig. 1, covering pipes up to 40 in in diameter and 30 ft long.

For the important region 1 μ and 1 mm pressure, where the flow changes from molecular to viscous, corrections have to be applied to the basic formulae. These have been reproduced in Fig. 2.

To illustrate the importance of gas flow information let us consider the case of a diffusion pump having a top flange of 8 in inside diameter and a speed of 1 000 l/sec. If the equivalent length of the connection to the vessel to be exhausted is 3 ft, the effective speed of the pump will be reduced to approximately 430 l/sec. In a practical design 3 ft is not a long connecting pipe, so it may be desirable to increase its diameter to 9 or 10 in to improve the overall efficiency of the pump.

TRIAL DESIGN

The object of the trial design is to determine the sizes and capacities of the major components. Those which are considered here are: the vessel to be exhausted, the pumps, the valves and the pipework. Having fixed at least approximately the size of the vessel it is then necessary to decide on the permissible time to exhaust it to the working pressure, and from this to select a suitable combination of pumps.

The size of the vessel will be determined by factors associated with the particular application and size of project, and is often outside the control of the vacuum designer. It is, none the less, important that there should be collaboration in the design, since a successful vacuum vessel is very dependent on correct manufacturing methods and techniques. Examples might be: the arrangement of welded and other joints, stress relieving and planned vacuum testing in the subassembly stages.

The choice of diffusion pumps cannot be determined by a simple calculation of the exponential law of exhaust, because in addition to making allowance for any special features of the process, the adsorbed gases on the walls of system have to be removed. In many cases the adsorbed layer of water on the metal surface represents the greatest delay in producing a high vacuum, and it is necessary to modify any calculations based on theoretical pumping speeds by a factor of 10 or more, to allow for the difficulty of removing this layer from the surface. If the walls of the vessel can be heated, or the humidity of air in the workroom can be controlled to a low value, the delay due to this factor is much reduced.

With allowances for the factors just mentioned a tentative design is made, based on the rate of pumping required and the ultimate pressure desired. The desired high vacuum pumping capacity may be supplied in the form of either a single pump of suitable displacement, or as a number of pumps in parallel or series parallel. Because of the physical size of the diffusion pumps their

efficient arrangement around the vessel to be pumped may have a major influence on the combination of pumps to be used. It is also very often necessary to consider the use of valves to isolate the diffusion pumps from the system so that they may be kept at working temperature while the product is removed and a fresh batch substituted. To obtain the maximum efficiency the connecting pipes, or manifolds, between the fine side of the diffusion pumps and the vessel should be short and of large cross-section. As a result, the valves must also be large and their physical arrangement in association with the pumps is part of the main layout problem.

The rotary pumps chosen have to be capable of rough exhausting the vessel in the available time, and also of handling the throughput of the diffusion pumps at, or below, the critical backing pressure. The characteristics of diffusion pumps are satisfactory for parallel operation, so it is not necessary to provide separate rotary pumps for each diffusion pump, and the fore vacuum pipes are normally joined into a common manifold, and thence via suitable isolating valves to the rotary pumps.

Before deciding on the number of rotary pumps for the system, it is advisable to consider the provision of a spare pump. This standby capacity is a useful insurance against failures which are sometimes due to contamination of the lubricating and sealing oil by vapours from the system.

CHOICE OF COMPONENTS

The vessel or tank. It is not possible to lay down hard-and-fast rules on how a vessel or tank should be constructed. Steel is probably the most common material because of ease of welding. Aluminium is possible and the continued improvement in aluminium fabrication techniques increases the interest in this material, particularly where a non-magnetic tank is required. Castings generally are to be avoided, although die-castings as used for small valve bodies are very reliable. The fabrication design should be planned on the drawing-board in such a way that as many welds as possible, if not all, can be tested separately in the construction stage. To do this it is necessary to provide interspaces either by arranging the runs of welding on either side of a plate, or by putting on a suitable permanent cover-plate of light gauge material purely as a testing fitment.

The testing will be in two stages, first sub-assemblies can have the leak rates on their interspaces checked and the leaks, if any, located and repaired. When the final leakage check is being taken on the completed vessel, it is usually possible to ignore the proved welds and to concentrate on the joints added in making the final assembly.

Valves. Valves designed specially for vacuum use are mostly plate valves with motion transmitted into the vacuum by means of a Wilson seal or one of its modifications. Tombac bellows are used and can be leak-free,

but since the permissible deflection of the corrugations is limited the gland assembly may be rather bulky.

Natural and synthetic rubbers are the materials generally employed as seating material and also as packing for flange joints. In special cases Silastic, Polythene and Teflon are used, but their relative lack of elasticity makes the valve design more critical.

Both hand and power methods of valve operation are widely used. In general, the operational efficiency of a plant is closely bound up with the ease of operation of the valves, and the author believes that the tendency is toward the power operation of valves. Available methods are: (1) a series of valves may be operated by mechanical linkages from a central camshaft; (2) electromagnetic operation in which a non-magnetic shield between the energizing coil and the plunger enables a glandless valve to be produced; (3) electric motor drives are convenient when short rated power units of small dimensions are available at an economical cost; (4) hydraulic and pneumatic methods use oil, water or air under pressure to move a piston in a cylinder giving a linear motion which is attractive for plate type valves.

For large plant pneumatic operation is perhaps the most widely accepted method. There is no great difficulty in making a neat design of valve to operate at a line pressure of 50–100 lb/in^2, and a positive pressure is maintained on the seating all the time the valve is closed.

Pipes and flanges. Pipework, other than the fine side connexions which are treated as part of the valve or tank design, can be made of welded or solid drawn steel tube. In the smaller diameters light gauge copper can be worth consideration, especially in view of the special trees, elbows, and other fittings which are available. Polythene pipe can be extruded in convenient lengths for handling, and welded after installation to form a continuous pipeline. For suitable applications this material may offer advantages to offset the high cost of raw material.

Flanges and the gaskets for them as applied both to pipework and components are a controversial subject. It would be of considerable benefit if standard sizes, bolt circles and gasketing methods could be agreed. In gaskets the essential difference of opinion is on the subject of single versus double gaskets. The author's own feeling is that for small systems and components, single gaskets are acceptable, but that for large pipelines double gaskets with facilities for testing *in situ* are the preferable solution.

SYSTEM CONTROL AND PROTECTION

Having determined the dimensions and approximate layout of the main components, and considered their design, the problem of achieving a high degree of operating efficiency remains to be tackled. In general, the operation of a plant is controlled by a number of valves, and certainly in the case of a large equipment an appreciable physical effort may be needed to operate

them. It is therefore often desirable to arrange for power operation of the valves, and this in its turn permits the control to be centralized at one convenient point where the necessary gauges may also be collected. In addition, the power operation of the valves allows some degree of protection against faulty operation of the system to be conveniently made.

The failure most likely to cause trouble is a rise of backing pressure in the diffusion pumps. This might be due to the development of an air leak in the system or to a power failure causing stoppage of the rotary pumps. If the diffusion pumps are permitted to boil at a high pressure, the working fluid may deteriorate at least temporarily, and may not be capable of producing the desired ultimate vacuum for several hours after the fault has been corrected. Further, if there is a large rush of air through the pump the operating fluid in its vapour form may be carried into the vacuum chamber or into the backing system, depending on the nature of

the failure, and will need to be replaced in addition to any cleaning of the vacuum chamber which may be necessary before the plant can again operate normally. It is, therefore, good practice to arrange for a vacuum relay to give warning of any undue rise of pressure, and by a suitable circuit arrangement to close valves and so isolate the pumps from the system. The pump heaters may also be interlocked with this same relay.

Building on this foundation, vacuum relays can be installed in strategic positions and the operation of valves interlocked to reduce the amount of skilled supervision needed for the operation of the plant.

ACKNOWLEDGMENT

Review of vacuum gauges

By W. STECKELMACHER, B.Sc., Research Laboratories, W. Edwards and Co. (London) Ltd., London, S.E.26

A short account is given of methods for measuring pressures below 10 mm of mercury and of the limitations of conventional gauges for accurate measurement. The applications of gauges to industrial processes and their use as protective devices are discussed.

In any process involving high vacuum technique it is usually necessary to know the gas pressure in the system. Since any particular gauge can only cover a small part of the whole pressure range there must be a large variety of gauges to cover all requirements. In order to decide the best method of measurement for any specific pressure range, it is necessary to consider which pressure dependent property of the gas is most affected, as well as its limitation on the accuracy of measurements. Generally, the pressure ranges can be sub-divided into: (a) high pressures, 760 mm–0·1 mm of mercury, where direct measurement of the pressure is possible, the force exerted by the gas being the operating factor. Below this range, the forces involved are too small for accurate measurements to be obtained. Gauges for these high pressures comprise manometers, capsule, and Bourdon type gauges of a large variety of designs. They will not be considered here. (b) Low pressures, 10 mm–10^{-8} mm of mercury, some pressure dependent property

of the gas is utilized. This may be Boyle's Law, heat conduction, viscosity, thermal molecular pressure or ionization. It must be appreciated that the measurement of these low pressures presents many difficulties, especially when the volume of the apparatus is small and the vacuum pipe lines are narrow. For accurate work any gauge will have to be connected to the vacuum system by wide bore lines offering the minimum of resistance to the gas flow, or capable of actual insertion into the system.

McLEOD GAUGE

This gauge was first developed in 1874 by McLeod; it is absolute and fundamental and is still extensively used as a reference gauge in vacuum work to-day. It is the only low pressure gauge which can be calibrated from its physical dimensions. As shown in the diagram, Fig. 1, a sample of the gas occupying the volume V and

at a low pressure p is compressed into a small capillary tube of cross-sectional area a by raising the mercury to beyond the cut-off. To decrease errors due to capillarity, the tube, AB, with the same cross-sectional area a is sealed on as a by-pass to the large pumping tube. One method of using the gauge is to raise the mercury in this
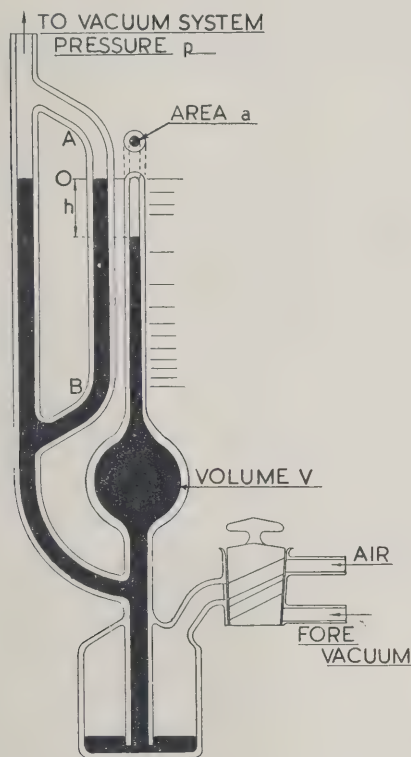
Fig. 1.   Diagrammatic sketch of the McLeod gauge

capillary to the same level as the top of the closed capillary. Then by Boyle's Law, we have

$$Vp = ah(h + p)$$

where h is the height of the mercury column supported by the compressed gas, p, being given in mm of mercury. V/a is the compression ratio of the gauge—a constant depending on the dimensions which determines the pressure range covered. There are naturally limitations to the value of this constant. For the lowest pressures V/a has to be as large as practicable; the limit for V is the weight of mercury to be handled and for a the sticking of the mercury in the capillary.

If the capillary tubes are made too small, slight differences in the surface conditions of the two measuring capillary tubes may cause large errors due to the different capillarity effects. It has been suggested that using larger diameter tubes this can be allowed for, to some extent, by viewing with a telescope and making an allowance calculated from the estimated contact angle of the meniscus. A sensitive gauge made in this laboratory has a volume of 400 c.c. and capillary of 1 mm

diameter. Values of 1 000 c.c. and 0·7 mm diameter respectively have been reported.[1, 2]

With these small bores the above capillarity difficulties are bound to arise and cannot be entirely overcome by the etching or grinding procedure sometimes recommended for the inside of the glass.

A McLeod gauge has recently been described in which a high compression ratio is obtained by fitting a small float valve at the entrance of the capillary, so that multiple compressions can be used. This allows the same sensitivity to be obtained as with a larger volume gauge.[3]

The McLeod gauge has some very serious disadvantages for general use. It is obvious that, if the system whose pressure is to be measured contains any condensable vapours such as water vapour, Boyle's Law is not obeyed and large errors may occur, though in special cases one can allow for this.[4, 5, 46] The operation is necessarily discontinuous and the operating fluid (mercury) has usually to be trapped from the rest of the system with cold traps. These traps do not remain perfect and may introduce further errors. Additional discrepancies may arise due to the surface conditions of the glass and mercury.

McLeod gauges using low vapour pressure oils as the operating fluid have been proposed, but introduce all the difficulties usually associated with oil manometer work; chiefly degassing, wetting of the glass with consequent time constant and possible hysteresis effect, etc.[6, 7, 8]

The McLeod gauge is, therefore, suitable only for use under very well-controlled conditions if accurate results are expected, the usual limit for consistent readings being about 10^{-5} mm of mercury.

THERMAL CONDUCTIVITY GAUGES

These gauges consist of heated wire filaments mounted in envelopes connected to the vacuum system. The thermal conductivity of the gas surrounding the filament is pressure dependent below 10 mm. At low pressures when the mean free path of the gas is of the same order or larger than the dimensions of the gauge, the energy loss by conduction in the gas is proportional to pressure, and also depends on the mechanism of the energy interchange between the solid surfaces of the wire and walls and the gas molecules, i.e. the accommodation coefficient of the surface, the structure and molecular weight of the gas. The heat lost by conduction through a gas of molecular weight M at pressure p between two surfaces at temperatures T_2 and T_1 is given by $Kap(T_2 - T_1)/(MT_1)^{\frac{1}{2}}$ cal cm^{-2} sec^{-1}, where K depends on the structure of the gas molecules and a is the accommodation coefficient.[8]

At very low pressure the energy lost by conduction becomes very small compared with the radiation loss, so that a natural limit for pressure measurements is about 10^{-4} mm of mercury.

The heated wires in these gauges may be operated under nearly constant energy input so that the temperature will decrease with increasing pressure. Different gauges employ different methods for measuring this change in temperature. This may be a thermocouple fixed to the wire (thermocouple gauge); or the wire may be made of high temperature coefficient material and its resistance measured by connecting it as one arm of a Wheatstone bridge (Pirani gauge). In both cases the meter may be directly calibrated in terms of pressure.

Thermal conductivity gauges have to be referred to other gauges such as the McLeod for their calibration. A factor of very great importance to the accuracy of the gauge is the surface condition of the filament. This may be improved to some extent by flashing the filament to a higher temperature before use.

In the Pirani gauge, the Wheatstone bridge may be balanced at atmospheric pressure (where thermal conductivity is independent of pressure) for sensitivity at higher pressures, or at a vacuum better than 10^{-5} mm of mercury (where almost all heat loss is by radiation) for sensitivity at low pressures. It has been shown that the optimum conditions for maximum sensitivity at a given pressure can be predicted.[9] The elements in the reference arms of the bridge should be similar to the elements in the measuring arms to ensure freedom from zero-drift due to ambient temperature changes. Fig. 2

Fig. 2. Pirani gauge showing the gauge head

shows an instrument of this type, the two reference arms of the bridge being evacuated and sealed off for a vacuum balanced gauge.

The variations of a length of wire with temperature, and the bending of a bi-metallic strip have also been utilized in thermal conductivity gauges.[10, 11] The former principle has been employed in this laboratory

successfully, in the form of a pressure sensitive relay, directly operated by the changing length of a fine wire of high thermal expansion alloy.

The thermal conductivity gauges have obvious applications in the medium vacuum range for control purposes and for the recording of pressures in vacuum process work as well as for leak testing. In addition, the large pressure range which may be covered and the ease with which all kinds of vapours are measured make them suitable for gas analysis[12, 13, 14, 15] and other chemical work.

VISCOSITY GAUGES

The effect of the gas surrounding a moving mechanical system is to cause damping depending on the viscosity of the gas. Like thermal conductivity this effect becomes pressure dependent at low pressures. To apply this principle to pressure measurements, a strip or fibre (which may be of quartz) is excited to vibration; the time for the amplitude of the vibration to die down to half its value can be taken as the measure of pressure (see, for example, Coolidge[16]). Alternatively forms of torsion pendulums using membranes, or disks of various forms, have been used and their performance has been studied by various workers (see, for example, Wetterer[17] and Dushman[52]). An interesting gauge combining the viscosity gauge at higher pressures with a form of Knudsen gauge at low pressures has been described by Gaede.[18]

In general the viscosity depends also on the molecular weight of the gas so that this gauge is specific for the gas or vapour in the system. This type of gauge has various advantages, which make it very suitable for some specialized applications in the pressure range 10^{-5} to 1 mm of mercury.[17] It can be made of small volume, is easy to outgas, and since it is a purely mechanical gauge which requires no heated parts, it is possible to measure chemically active vapours and does not suffer from contamination or clean-up.

IONIZATION GAUGES

There are several ways in which the ionization property of a gas can be utilized for measuring low pressures. At higher pressures the self-maintained glow discharge between two electrodes is extensively used. One can obtain an indication of the order of pressure in the system by direct observation of the glow, or the discharge current at a certain applied voltage may be measured or used for control purposes in connection with relays (see, for example, Bainbridge and Sherr[19]).

The glow discharge cannot be maintained at low pressures in the ordinary way except by increasing the voltage to a much higher value. The Penning or Philips gauge gives an extension of the self-maintained glow discharge into the low pressure region by increasing the average electron path and, consequently, the probability

of collision and ionization. This is achieved by employing crossed magnetic and electric fields. Various electrode configurations have been employed (see, for example, Penning,[20] Makinson and Treacy,[21] and Garrod and Gross[22]) which, with suitable magnetic field, force the electrons to oscillate in helical paths. The collision probability is so increased that large discharge currents can be obtained at reasonably low voltages and small electrode distances. A gauge of this type is illustrated in Fig. 3. It consists of two parallel
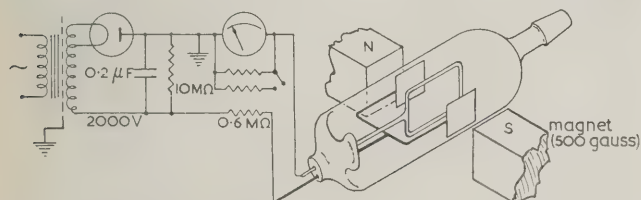
Fig. 3. Showing circuit and gauge head of Philips gauge

plate electrodes about 1 in apart as cathodes symmetrically placed on opposite sides and parallel to an anode ring. The magnetic field is perpendicular to the cathodes and axial to the anode ring. With a magnetic field of 500 gauss and a conventional 2 000 V power supply, the gauge is sensitive over the range 5×10^{-3}–10^{-5} mm of mercury. A typical calibration using a 1 mA meter is shown in Fig. 4. By altering the electrode
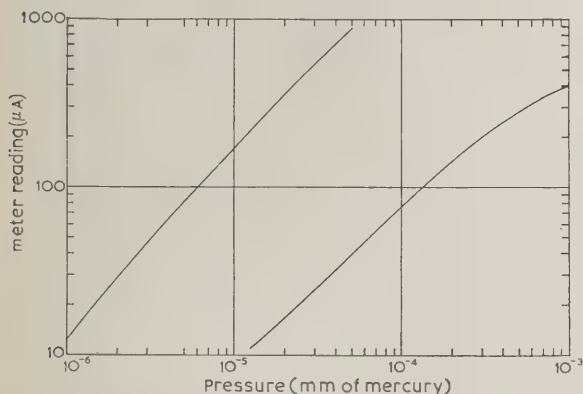
Fig. 4. Calibration of Philips gauges connected as in Fig. 3. The lower curve refers to a gauge with ring anode and a cathode spacing of 2·5 cm. The upper curve refers to a gauge with cylindrical anode and a cathode spacing of 6 cm

configuration various calibration characteristics can be obtained.[23, 24, 25] It is found that the calibration depends also on the gas in a similar way to thermionic ionization gauges [26, 27] (see also below).

A gauge of similar design to the original but of much larger dimensions (cathode distance 6 cm) using a cylindrical anode and a magnetic field of 450 gauss has been constructed in this laboratory. This gives greatly improved sensitivity and an almost linear calibration at

low pressures. Thus at a pressure of 10^{-6} mm of mercury at 2 000 V the current was 12·5 μA.

This important development will have many applications for low pressure work where it has the advantage over the thermionic type of ionization gauges in view of the absence of fragile filaments. The life of the gauge is consequently increased. An important application was described by Penning and Nienhuis[25] recently, in
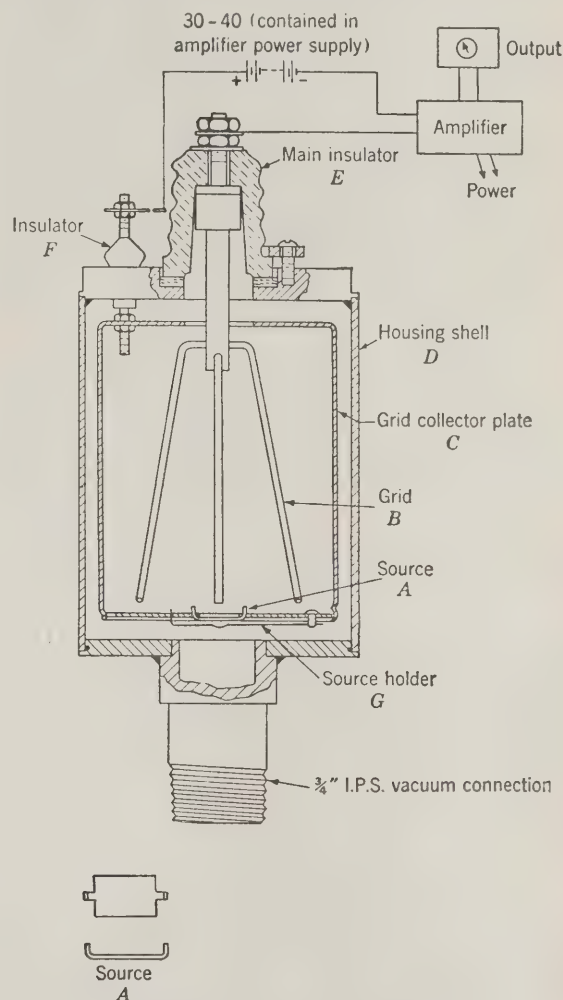
Reproduced from Rev. Sci. Instrum.

Fig. 5. Alphatron gauge head

which a gauge of this type was sealed off at low pressure with a palladium barrier. This has been used as a sensitive leak detector with hydrogen as the searcher gas.

For the higher pressure range (from 10^{-3}–10 mm of mercury), a form of ionization gauge, the Alphatron, has been designed containing a capsule with a minute quantity of radium, the radiation from which ionizes a small percentage of the gas in the gauge head. The ionization is produced by alpha particles and the gauge head consists essentially of an ionization chamber and source as shown in Fig. 5.[28] The source is shown at A,

and 40 V are supplied between grids *B* and *C*, so that the very small ion currents are collected and can be amplified by a very sensitive d.c. amplifier. The ionization currents produced are of the order of $2 \cdot 5 \times 10^{-10}$ A per mm of air pressure. In a gauge of this type, the calibration is linear and with 3-range switching one can get $0 \cdot 1$, 1, and 10 mm of mercury full-scale deflexion on a 200 μA meter. Calibration curves showing the effect of different gases are shown in Fig. 6.
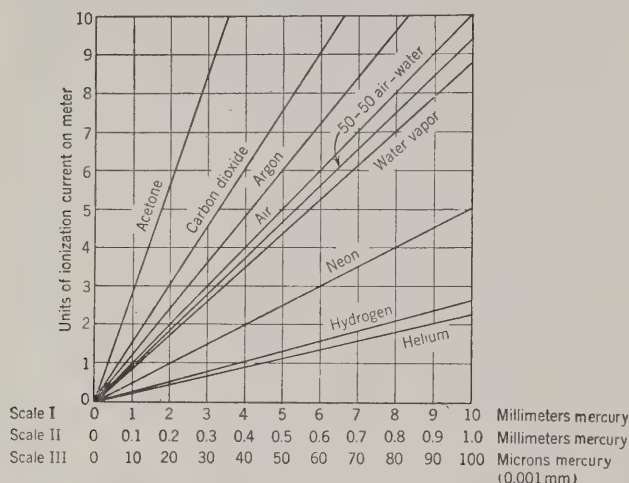
Fig. 6.—Calibration of an Alphatron gauge

An advantage of this gauge is the linearity of the calibration over this pressure range. As disadvantages one should mention the precautions necessary to avoid any possible poisonous effects from the radium emanation (as discussed in the operating instructions supplied by the manufacturers).

Another form of ionization gauge makes use of the electron emission from a hot filament. The emission may be temperature limited and kept at a constant value so that the ionization by collision with the gas is proportional to the gas density. The number of positive ions formed is measured by attracting them to a third negatively charged electrode and determining the current required for neutralization. The pressure is measured by virtue of the equation

$$p = kc \, i^+/i^- \tag{3}$$

where i^+ represents the ion current (μA) and i^- represents the electron current (mA), p is the pressure in mm of mercury, k is a function of the potentials and the geometry of the gauge and c is a factor depending on the relative probability of ionization of different gases referred to air.[26] It is seen that the calibration is linear with fixed potentials and constant emission for a certain gas so that, theoretically, a single calibration point would be sufficient for all pressures.

Different constructions have been used, the usual

voltages and currents ranging from 100 to 500 V on the electron collector, 10 to 30 V (negative) on the ion collector and emission from $0 \cdot 1$ to 20 mA respectively. Holloway[26] has summarized different constructions.

For many practical purposes where a continuous reading of pressure conditions is desirable special circuits have been designed with the purpose of maintaining the electron current i^- constant automatically. This is necessary as it is found that a gauge operated with a steady filament current still gives wide fluctuations in emission owing to filament poisoning effects, temperature variations, etc. Many different circuits have been proposed[29–32]; one will be described briefly below. The ion currents obtained with any thermionic ionization gauge are very small so that if sensitive galvanometers are to be avoided, the control unit must incorporate a d.c. amplifier which can be of conventional design, such as those described by Bowie[33] and Krusser.[34]

A typical circuit of a control unit for stabilizing the emission is shown diagrammatically in Fig. 7. The filament current is supplied by a transformer which is fed partly through a thyratron. The thyratron obtains its bias from a resistance carrying the electron current of the gauge. The thyratron is firing or quiescent in opposition to emission fluctuations supplying less or more filament current accordingly. The switching action may be observed on a monitor lamp. This method ensures a constant electron current under varying pressure and filament conditions.[31]

The range of pressures covered by conventional thermionic ionization gauges is very large, extending
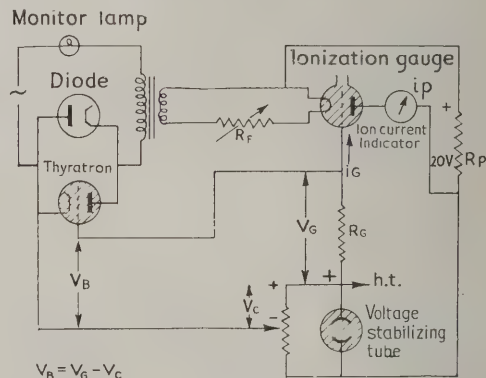
Fig. 7. Control circuit of ionization gauge for emission regulation

from about 10^{-2} to 10^{-7} mm of mercury. At higher pressures the linearity falls off, whereas the lower limit seems at first to depend only on the sensitivity of the ion current measurement and should be able to be extended into the 10^{-9} mm region. At the low ion currents expected at these pressures it is important carefully to eliminate leakage currents on the glass surface, photoelectric effects, etc., which may become

comparable in magnitude. Even then another un-expected effect has been observed. Investigations on oxide-cathode valves by Metson have shown that there is a lower limit (depending on the accelerating voltage used) attributed to the release of electrons from the ion collector by soft X-rays generated at the surface of the bombarded electrode. This results in a spurious ion current.[35-37]

Fig. 8. Ionization gauge heads with "fast" and "slow" tubulation

The thermionic ionization gauge suffers from several disadvantages more or less serious depending on its particular application. The presence of a filament at a high temperature may cause dissociation of the gas or vapour to be measured. This may cause poisoning of the emissive surface and lead to filament burn-out. Gauges have been made with tungsten, tantalum and oxide-coated platinum or nickel filaments, all of which suffer from these effects.[38]

The other disadvantage is that, in common with all apparatus utilizing a gas discharge, there are electrical clean-up and degassing phenomena. This leads to dynamic conditions in the gauge head which may seriously affect the pressure readings.[39, 40, 41] Further-more, the exact composition of the gases in the gauge head is generally not known since there will be gases evolved from the walls of the system and decomposition may be taking place at the hot filament. Consequently, there is always a degree of uncertainty as to the correct ion constant to be taken. (In sealed-off devices the pressure is often referred to indirectly by giving the value of the residual ion current.)

It has been said by Schwarz, who has made an investi-gation of these effects, that ionization gauge readings below 10^{-4} mm of mercury should be regarded with some suspicion unless special precautions are taken. The effect can be minimized by avoiding, if possible, the flow resistance offered by restrictive tubulation between the gauge head and the point at which the pressure is to be measured. Fig. 8 shows a range of ionization

gauges with "fast" connexions of wide bore tubing and a gauge for mounting directly into the system.

Both the thermionic and the Penning type of ionization gauge have found very extensive applications for measuring the highest vacua in the laboratory. In applications to electronics one is usually only interested in the ratio of ion current to the ionizing electron beam. Some of the difficulties mentioned above may not then arise.

Another application which is very important for vacuum work in general is the use of ionization gauges for leak detection. For this purpose, the gauge has been modified by separating it from the vacuum system by a small palladium window. The palladium may be heated either by a separate heater or, in the case of the ther-mionic gauge, by bombardment by electrons. In this condition, the palladium is permeable to hydrogen only so that small changes in concentration of this gas are detected by the gauge.[25, 42]

KNUDSEN GAUGE

At low pressures when the mean free path of the gas is long relative to the distance between two plates at temperatures T_1 and T_2 in an enclosure at tempera-ture T_2, it has been shown by Knudsen that there is a repulsion between the plates which is proportional to the gas pressure p.

Molecules leave the hot plate (at T_2) at a greater mean velocity than molecules from the colder walls (at T_1) of the gauge chamber and transport more momentum to *one* side of a suspended vane. This is the principle underlying the operation of the Knudsen gauge, various forms of which have been described by Klumb and Schwarz.[43] In one of these the vane is suspended axially in the vertical plane as shown in Fig. 9. The
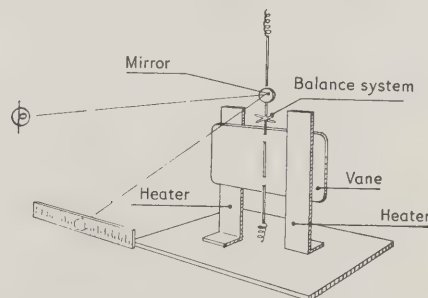
Fig. 9.—Diagrammatic sketch of Knudsen gauge

hot plate takes the form of two heaters placed on diametrically opposite sides of the vane to increase the repulsive effect. The deflexion θ experienced by the vane (at the pressure p) is given by

$$\theta = \tfrac{1}{2}pk[(T_2/T_1)^{\frac{1}{2}} - 1] \qquad (4)$$

where the constant k depends only on the geometry of the gauge. This formula assumes that the accommoda-tion coefficients of heater and vane are unity. Allowing

for the accommodation coefficient, it is found that for low temperatures the correction is of second order and, therefore, the effect of different gases on the calibration can only be of this order.[43, 44] It has been found in practice that calibrations for different gases are within 10% of each other.

In the derivation of equation (4) one could take into account the finite size of the vane, the distance between the heater and vane,[45] and the torsional constants of the suspension to calculate the constant k from the

Fig. 10. Commercial form of Knudsen gauge

dimensions of the gauge. In practice it is more convenient to calibrate against another gauge.

The pressure range covered by a Knudsen gauge is necessarily very limited as the only variable parameter is the temperature of the heaters. It is possible by using different suspensions to cover the range of 10^{-2} to 10^{-7} mm of mercury. At the higher pressures the deflexions become non-linear with pressure as the radiometer region is entered.[46] At lower pressures one

T_1 as low as possible for high sensitivity, although this is of considerable practical difficulty.

A suspended system of this kind in a vacuum is usually very sensitive to vibrations. The effect can be much reduced by the use of a taut suspension together with magnetic damping.

A modern Knudsen gauge designed along these lines is shown in Fig. 10. The deflexions are observed by a lamp and scale as in a mirror galvanometer. The vibration sensitivity is small so that the gauge may be bolted directly to the vacuum system by the large-bore flanged pumping connexion. In this case the two ranges, corresponding to two heater temperatures, are 10^{-4} and 2×10^{-5} mm of mercury full scale deflexion on a 10 cm scale using a 20 cm scale distance. The calibration is substantially linear.

It is clear that the Knudsen gauge is the only gauge in this pressure range the sensitivity of which is not affected by vapours and different gases, and hence it should find many applications in the whole field of high vacuum technology. This was first pointed out by Du Mond,[47] and recently a Knudsen gauge has been proposed for measuring vapour pressures.[48]

CONCLUSION

In this review it may seem that a disproportionate space has been given to details and applications of the McLeod gauge. But this is our only really absolute manometer serving as the standard gauge, all other gauges ultimately depending on it for their calibration. The difficulties that may arise in its use and its limitations for practical measurements have been pointed out, and for these reasons the various other gauges have been developed so that the whole range of vacuum measurement may be covered as shown in Fig. 11. Fortunately there is sufficient overlapping to allow selection of a suitable gauge for any particular application.
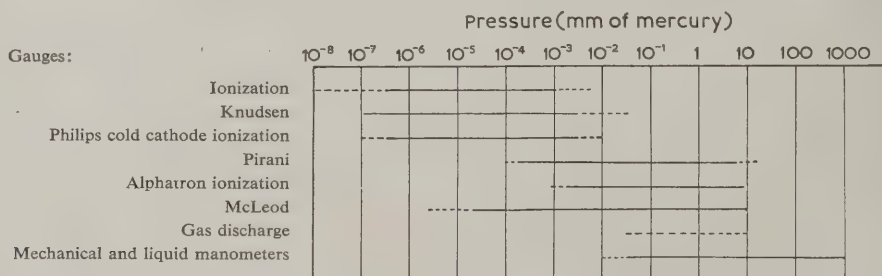
Fig. 11. Pressure "spectrum" of vacuum gauges to show the ranges covered by some of the principal ones
Full lines indicate range of greatest sensitivity and reliability

could use more delicate suspensions, but it can be shown that the thermo-molecular forces become comparable in magnitude with the radiation pressure from the heater. At pressures of 10^{-6} mm of mercury the radiation pressure may account for 5%–10% of the total force on the vane and, at 10^{-8} mm of mercury, it may account for as much as 80–90%. It is advantageous to keep

In practice one has to consider such things as the interaction between the measuring instrument and the gas in the system. The gas or vapour in the system may be corrosive and limit the choice of materials with which it may come in contact, thus even normal gases and vapours may damage essential parts of a gauge, e.g. the poisoning of ionization gauge filaments by fairly

low concentrations of hydrocarbon and water vapours. On the other hand, the mercury or oil vapour of gauges requiring these liquids may be interacting with the rest of the system. The presence of heated parts may lead to the thermal or catalytic decomposition of some of the gas in the system; temperature differences in a gauge, or in the gauge relative to the system, may cause distillation or condensation. There are gettering or gas clean-up effects which may have to be considered, especially with gauges employing the ionization principle, while the outgassing of the walls of the apparatus as well as the free surfaces inside the gauge represent special problems in many cases.

In some applications the means of recording the pressure or pressure changes may be of importance and one may then have to consider the response time of the instrument and the possibility of indications or recordings remote from the vacuum system.

Another problem which sometimes arises is that the gauge calibration may be specific for the gas or vapour in the system and hence a knowledge of the composition of the gas may be necessary to interpret the gauge readings. One can see that there are many applications in which this knowledge is not readily available.

We can therefore see that there is a whole range of factors which has to be considered with vacuum measurements before the selection of the most suitable gauge for any particular application can be made.

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DISCUSSION

Mr. C. F. Machin: In connexion with the Knudsen gauge, Mr. Steckelmacher referred to the fact that at very low pressures difficulty is sometimes experienced with continued oscillation of the movable vane through lack of viscous damping of the vane by the gas. At higher pressures this effect can, of course, be used as the basis of a mechanical gauge of the oscillating quartz fibre type. Here the logarithm of the time of decay to half amplitude is a more or less linear function of the logarithm of pressure over the range of 10^{-3} to 1 mm of mercury.

The gauge can only be used where the pressure in the system does not change rapidly with time. It has, however, particular application to the measurement of the pressure of an active gas such as hydrogen for there is neither contamination nor clean up.

Author's reply: I only briefly mentioned gauges of the viscosity type. An interesting gauge using the viscosity principle at higher pressures and the Knudsen principle at lower pressures has been described by Gaede.[1] Alternatively, torsional pendulums using fibres, membranes or disks of various forms have been used.[2] A detailed analysis of the performance of quartz fibre viscosity gauges has been made by Wetterer.[3]

Dr. D. G. Marshall: Why was the Philips gauge described in the lecture contained in a glass envelope? Would it not be an advantage if more ionization gauges were manufactured with metal envelopes instead of the conventional glass one?

Author's reply: It was purely a matter of convenience. In designing a gauge with a metal envelope special care would have to be taken to prevent electrical leakage on the insulated high-voltage lead. Also degassing problems from the gauge envelope would have to be watched, especially in the ionization gauge because of the heat dissipation. Water cooling of the metal envelope may be desirable. There are many applications where metal envelopes would be an advantage.

Dr. M. Pirani: The degassing of ionization gauges, by heating the filament to a high temperature and applying a high voltage to the anode, damages the filament, especially when hydrocarbons are present. Therefore, British American Research Ltd. use a gauge construction in which the degassing is achieved by passing current through the grid (a tungsten spiral) and raising the temperature gradually to 800° C, thus using it as a "furnace winding." The filament is not switched on before a vacuum of 1 μ is reached and it is never overheated.

Author's reply: I would like to refer again to the earlier remarks regarding the dangers of obtaining dynamic conditions in the ionization gauge, especially after degassing procedures, however they may be carried out. There are, however, applications when degassing may be an advantage, as, for instance, when using the gauge for different gases.

Mr. A. D. LeClaire: Mr. Steckelmacher mentioned that by suitable attention to design, the range of operation of the Pirani gauge may be extended up to 10 mm. It is unfortunate that so little attention seems to have been given to the development of conductivity gauges for use in this pressure range for there is no other gauge so suitable for use at such pressures in apparatus which is to be baked and sealed off.

The method of construction required for this extension of range seems to be a matter of some dispute in the literature, although the physical principles involved are surely well known. Some writers hold that it is necessary to make the wire to wall distance as small as possible, while others maintain that the most important requirement is a very fine wire and that the wire to wall distance is of secondary importance. All agree, however, on the importance of running the wire at a higher temperature than is normally employed and of using a balanced bridge method of control so as to maintain this required higher temperature as the pressure rises. It is surprising that commercial manufacturers claim for their Pirani gauges a range of operation up to 10 mm and so often pay little attention to these points of construction and operation.

Mr. Steckelmacher has brought to my notice a series of papers by H. v. Ubisch[4] which deal quite thoroughly with the theoretical and practical aspects of Pirani gauge operation. The conclusions are that the upper limit of usefulness of the gauge is inversely proportional to the diameter of the wire, that the range increases "somewhat" with increasing wire temperature and that since "the dimensions of the enclosing bulb enter logarithmically" into the expression for the heat conducted away from the wire "they have no significant influence as long as

they are large compared with the diameter of the wire, which is the most critical factor." The effect of closely spiralling the wire is to decrease the sensitivity and at high pressures the length of the wire is of less importance than at low pressures.

That the only essential requirements for the extension of the range of a conductivity gauge into the higher pressure region is a reduction in the wire diameter is well confirmed by the experiments of Knudsen,[5] Bartholomeyczyk,[6] Kersten[7] and Rittner[8] as well as of Ubisch.[4] Rittner implies in his paper that his success was due to a small wire–wall distance, but he fails to point out that he used a very fine wire (0·01 mm). Ubisch compared the calibration curves of two gauges which were identical except that their bulb diameters were, respectively, 2·6 cm and 0·23 cm and found them to "correspond almost exactly."

Author's reply: I would like to endorse the remarks made by Mr. Le Claire. As an example of the misleading statements referred to one might mention a very good recent book on vacuum technique.[9] The correct situation may be deduced from considerations of the kinetic theory of gases, by which the heat conduction between two concentric cylinders may be calculated (see, for example, Kennard[10]) from which the above-mentioned results follow.

Dr. M. Pirani: Hot wire gauges can be used up to atmospheric pressure when convection is used.

Mr. I. Varnam. The inaccuracy of the McLeod gauge in the presence of condensable vapours has been discussed. It is felt that mention should be made of methods of overcoming this difficulty which permit the true measurement of total pressure and partial vapour pressure.[11]

Dr. M. Pirani: The McLeod gauge can be used for measuring vapour pressures, e.g. water vapour in air, by taking readings at different compressions. Corrections for vapours which are present in air have been given by Melville and Farkas.[12]

Mr. G. A. Carrick: We once made a McLeod gauge which we hoped would be very sensitive, with a bulb of volume 500 c.c. and a tube of about 1 mm bore. Successive readings with this gauge increased uniformly, e.g. 0·1, 0·2, 0·3, up to 1 μ or more. There was not, in this case, any pellet of mercury at the entrance to the tube and, further, the same size of tube used with a smaller bulb did not show this effect. Has the author any experience of similar results?

Mr. N. Ochert: I would like to suggest that a probable cause of this effect was a film of air between the mercury and the glass in the bulb or tubing below the bulb. If, as in the early McLeod gauges, a rubber tube connected the mercury reservoir to the gauge proper, this is another possible cause and an air trap, such as may be observed on the modern commercial forms of McLeod gauge just below the cut-off point, is essential.

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General principles of leak detection

By J. Blears, B.Sc., A.M.I.E.E., Metropolitan-Vickers Electrical Co. Ltd., Manchester,
and J. H. Leck, M.Eng., Electrical Engineering Department, University of Liverpool

The techniques of leak detection are reviewed, special emphasis being placed on methods for obtaining high sensitivity with simple apparatus. A theoretical analysis of the gas flow problem is supplemented by experimental results obtained with a system using two diffusion pumps in cascade. The importance of using constrictions and correctly positioning the gauge is stressed, and the losses of sensitivity due to virtual leaks, leaks in series and by-pass methods are considered. The relative sensitivities of different leak detection methods are tabulated.

1. INTRODUCTION

The aim of this review is to present an overall picture of the principles of leak detection in vacuum systems. Emphasis will be placed on the simpler methods because the techniques using mass spectrometers have already been extensively reviewed.[1,2,3,4] An attempt will be made to show how the simpler methods, using readily available instruments, can be operated with a sensitivity adequate for all leak detection problems encountered in the operation of dynamic vacuum systems. Compression methods, e.g. the halide leak detector[5] or the soap bubble method[6] will not be described. The general method to be considered is that in which the position of a leak is determined by detecting the presence *inside* the vessel of a gas or vapour sprayed on the *outside* whilst the vacuum system is operating under normal pumping conditions. This jet of gas or vapour is often referred to as the "probe gas," or more simply as the "probe."

Since the advent of vacuum engineering the gas discharge tube has been the best known and most commonly used instrument for leak detection. It was not until after 1930 that more sensitive and reliable methods were described. In 1935 T. E. Cuykendall[7] pointed out the possibility of using a Pirani gauge in the fore vacuum line in conjunction with a coal gas probe. The rapid expansion in vacuum engineering associated with the atomic energy programme in the early 1940's led to a scientific investigation of leak detection. In Great Britain, work was concentrated on the improvement of existing instruments for measurement of small changes in gas pressure and composition. This work is described in a series of reports written in the years 1941–45.[8] In the U.S.A. similar researches carried out by the Kellex Corporation were reviewed by Jacobs and Zuhr in 1947.[1] These authors concluded that the mass spectrometer, with helium as a probe gas, was by far the most sensitive and useful instrument for leak detection purposes and much of their review is devoted to the use of this technique.

The position to-day is that the mass spectrometer leak detector has been extensively developed both here and in the U.S.A.[9] It is now an efficient and certainly an extremely sensitive leak detector and Jacobs and Zuhr's conclusion that this instrument will always be preferred where its use can be justified economically is generally accepted. There are many applications where such justification does not exist; consequently, there is a need

for simpler techniques where the sensitivity, though not as high as that of the mass spectrometer, is still adequate for most purposes. This need has led to the development of the halide leak detector in which the normal "vacuum" method is reversed. The vessel to be tested is filled with a halogen gas (such as Freon), and the leak is then found by detecting the presence of the halide outside the test vessel.[10] Other recent developments include the so-called differential methods which will be described below in section 3.3.

Little has been published on the methods for obtaining optimum sensitivity with the simpler vacuum techniques and it is hoped that this paper will to some extent remedy this deficiency.

2. DETECTORS

The instruments listed in Table 1 are commonly used to indicate the presence of the probe gas in the vacuum systems:

Table 1. *Instruments used to indicate presence of probe gas*

	Maximum useful operating pressure (microns)	Minimum detectable pressure change (microns)
Geissler tube	10^4	10
Penning gauge	1	10^{-5}
Ionization gauge	10^{-1}	3.10^{-7}
Mass spectrometer	10^{-2}	10^{-8}
Diode	10^{-1}	10^{-5}
Pirani gauge	100	10^{-3}

2.1 *Ionization gauges.*—The Geissler tube is too insensitive for any but the grosser leaks but, as most other detectors have an upper limit beyond which their sensitivity falls to a small value, the Geissler tube is often used in practice to detect leaks which give rise to pressures greater than 100 μ.

Both the Penning gauge and the ionization gauge depend for their action on the fact that probe gas molecules may have a different ionization efficiency from that of air. Thus, by choosing a probe gas with the right ionization constant these gauges can be made to form more (or less) ions when the leak is covered and, thus, the leak can be detected by observing the changes of ion current following application of the probe. The difference between these gauges and the Geissler tube lies in their enormously greater sensitivity, comparative minimum detectable pressure changes being as tabulated

above. To obtain the sensitivity tabulated, the Penning gauge must be of the sensitive type, such as that described recently by F. M. Penning and K. Nienhuis.[11] This gauge, which has a cold cathode and therefore cannot be burnt out, operates over the pressure region 10^{-4} to 1μ. The hot filament gauge, whilst being liable to burn out if not protected by a vacuum relay, is rather more stable. For example, with an automatically controlled gauge using the Ridenour-Lampson circuit[12] it has been found possible to detect a pressure change of $3 \times 10^{-7} \mu$ against a base pressure of $3 \times 10^{-4} \mu$. This 0.1% limit was imposed by fluctuations introduced by the control circuit.

2.2 *Gauges depending on work function.*—The decrease in electron emission from a tungsten filament when contaminated by oxygen has been used to detect changes in the oxygen concentration in vacuum systems when this gas is used as a probe.[13] R. B. Nelson[14] has described a control unit which automatically maintains a diode filament at a constant temperature. The control is effected by running the diode filament in series with the filament of a similar sealed-off diode, the latter being forced to give a constant emission by means of the Ridenour–Lampson regulator circuit. This principle has been extended by Riddiford who used a three-electrode ionization gauge, operated at constant filament voltage, and took advantage of the increase in electron emission caused by covering the leak with an organic
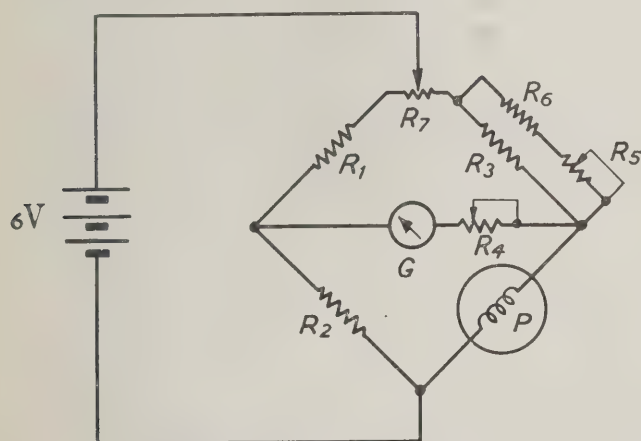
Fig. 1. Circuit diagram for Pirani gauge control unit. 6 V accumulator used for power supply. Pirani gauge operating temperature 100° C. Galvanometer G, 450 Ω 170 mm/μA. P = Pirani element; R_1, R_2, R_3 = 100 Ω; R_4 = 50 kΩ; R_5 = 5 kΩ; R_6 = 5kΩ; R_7 = 15 Ω

solvent such as tri-chlorethylene. By choosing a gas with a high ionization efficiency (probability of ionization) and a marked lowering effect on the work function of tungsten, the advantages of both the diode and the ionization gauge can be combined. This is because the reduction of work function increases the electron emission (and therefore the gauge sensitivity) while the high

probability factor increases the positive ion current still further.

However, there are always other gases and vapours present in vacuum systems[15] and any time changes in the concentrations of these gases will produce consequent changes in work function. All "diode" systems are thus very prone to drift and a considerable time (15 min to several hours) must always elapse before they can be used with full effect. In our experience one of the most encouraging features of gauges depending upon work function is the fact that once equilibrium has been established a change of gas composition produces a well-defined, rapid and reversible change of electron emission. Even with total pressures as high as one micron a new equilibrium value of emission is established within a second of changing the gas composition. Recovery after removing the probe is slower, but seldom takes longer than 15 sec.
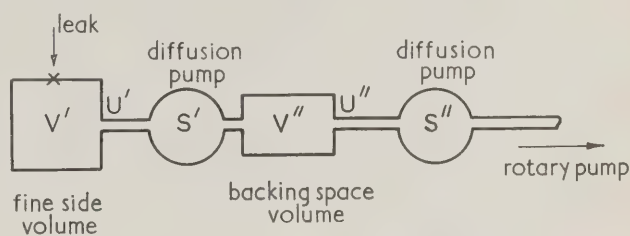
Fig. 2. Typical pumping plant. Constrictions U' and U'' are made variable so that pressures or time constants may be adjusted to give optimum sensitivity

2.3. *Thermal conductivity gauges.*—The Pirani and thermocouple gauges can be used for leak detection because gases with thermal conductivities different from that of air can be used as probes. Although thermal conductivity gauges are much less sensitive than ionization gauges they can be operated at pressures 100 to 1 000 times higher and, furthermore, they have the advantage of robustness and simplicity of control which make them very suitable for operation by relatively unskilled labour. With the circuit shown in Fig. 1 and using a 3 in layer of cotton wool to protect the gauge from draughts, gauge instabilities give rise to apparent fluctuations of approximately $10^{-3} \mu$, i.e. a sudden pressure change of $10^{-3} \mu$ can just be detected. Very much greater stability can be obtained by using compensating gauges with envelopes at liquid nitrogen temperature. Knauer and Stern,[16] for example, quote a stability of 10^{-5} to $10^{-6} \mu$, but such performance can only be attained by taking most extraordinary care. This work, however, does suggest that a performance considerably better than the accepted $10^{-3} \mu$ might be obtained by operating with the gauge envelopes at $-78°$ C.

3. FLOW SYSTEMS

Whilst it is quite obvious that the detector element is the heart of any leak detection system, the design of the

associated vacuum plant and the positioning of the detector element in this plant are factors of equally great, and sometimes even greater, importance.

Probing a vacuum system gives rise to pressure changes which are transient in nature, and a full mathematical treatment involves the differential as well as the steady state solutions of the flow equations. The latter, which are simpler, will be treated more fully than the former.

3.1. *Steady state pressure equations.*—In the following sections the symbols used are:—

V = volume.

U = effective speed of pump and its series constriction.

P = pressure.

L = leak rate.

M = molecular weight.

η = coefficient of viscosity.

x = fractional concentration of probe gas in atmosphere.

$'$ $''$ are superscripts denoting "fine side" and "backing side."

A, P, x are suffixes denoting "air," "probe gas," "air/probe gas mixture."

x', s = particular values of x ($x = 0 \rightarrow 1$) ($s = 1$).

K_A, K_P = gauge sensitivity constants.

G = gauge deflexion.

Δ = fractional change.

ϕ = ratio (apparent change of air pressure caused by covering leak with probe)/(mean air pressure before probing).

A typical pumping plant is shown in Fig. 2. This plant consists of a test vessel of volume V' l continuously evacuated by two diffusion pumps and a rotary pump in cascade. The effective pumping speeds of the diffusion pumps are reduced by constrictions to U' and U'' l/sec. The volume between the two diffusion pumps (backing space volume) is V'' l. (This volume includes that of the gauge or other leak sensing element.) Corresponding air pressures are P'_A and P''_A and the equation of continuity states that in the steady state the leak rate is

$$L_A = U'_A P'_A = U''_A P''_A \tag{1}$$

The flow through the leak itself may be diffusive or viscous, or it may be a combination of both. Taking as a criterion for diffusive flow the fact that the mean free path should be equal to or greater than the diameter of the leak, the maximum diameter for pure diffusion is about 5×10^{-6} cm and the corresponding maximum leak rate for a hole 2 mm long would be 10^{-8} to 10^{-9} μl/sec. It is thus apparent that for the more common leaks, i.e. those between 10^{-4} and 10 μl/sec, the flow through the leak itself must be viscous. Whichever is the case, however, the total flow is given by the Knudsen equation.[17]

$$L = \left(A \frac{1 + cPM^{\frac{1}{2}}\eta^{-1}}{1 + fPM^{\frac{1}{2}}\eta^{-1}} + B\frac{P}{\eta}\right)P \tag{2}$$

where P is the total pressure applied to the leak; A. B. c and f are numerical constants depending on the dimensions; M is the molecular weight and η the viscosity of the gas. We shall consider only the practical case where the flow through the leak is entirely viscous and assume the probe gas concentration over the leak is x atmospheres, the air concentration being $(1 - x)$ atmospheres ($x < 1$). Equation (2) then simplifies to

$$L = \frac{B}{\eta}P^2 \tag{3}$$

The essential characteristics of a viscous leak are: (a) that it is non-fractionating, i.e. that a probe gas concentration x, outside the leak, will result in a fraction x probe molecules entering the vacuum system; (b) that the total flow is determined by the inverse viscosity of the gas *mixture* and the square of the total pressure. (Since the square of the total pressure is constant, P^2 has been omitted from the following analysis.) Combining equations (1) and (3):—

$$L_x = B/\eta_x = U_A P_A + U_P P_P \tag{4}$$

where the suffix x refers to the mixture of air and probe gas. From the leak characteristics (a) and (b) it follows that:

$$\begin{aligned}(B/\eta_x)(1 - x) &= U_A P_A \\ (B/\eta_x) . x &= U_P P_P\end{aligned} \tag{5}$$

Hence the total pressure inside the vacuum system is

$$P_A + P_P = \frac{B}{\eta_x}\left(\frac{1 - x}{U_A} + \frac{x}{U_P}\right) \tag{6}$$

The change in pressure at the gauge when x changes from zero (probe absent) to a specific value x' (leak partly covered by probe gas) is:—

$$\Delta P_{x'} = B\left[\frac{1}{\eta_{x'}}\left(\frac{1 - x'}{U_A} + \frac{x'}{U_P}\right) - \frac{1}{\eta_A U_A}\right] \tag{7}$$

The maximum change in pressure when $x = 1$ is:—

$$\Delta P_S = B\left(\frac{1}{\eta_P U_P} - \frac{1}{\eta_A U_A}\right) \tag{8}$$

The pressure is read by means of a gauge having a sensitivity K_A for air, K_P for the probe and by substituting $G = KP$ in equations (6)–(8) we find the corresponding gauge deflexions. Equation (8), for example, gives:—

$$\Delta G_S = B\left(\frac{K_P}{\eta_P U_P} - \frac{K_A}{\eta_A U_A}\right) \tag{8a}$$

From this equation the requirements for maximum change in gauge reading following probe substitution can be seen clearly. They are: (1) complete coverage with the probe gas; (2) high gauge sensitivity for the probe gas; (3) low probe gas viscosity; (4) high molecular weight (since $U \propto M^{-\frac{1}{2}}$); (5) small value of the conductance U. Probe gases are considered in Section 3.4.

The constant most under the control of the plant designer and user is the value of the conductance of the constriction. This value should be low, but it cannot be reduced indefinitely, the limits being imposed (*a*) by the maximum permitted gauge pressure (Table 1) and (*b*) by the time required to change the atmosphere inside the vacuum system after the probe gas has covered the leak.
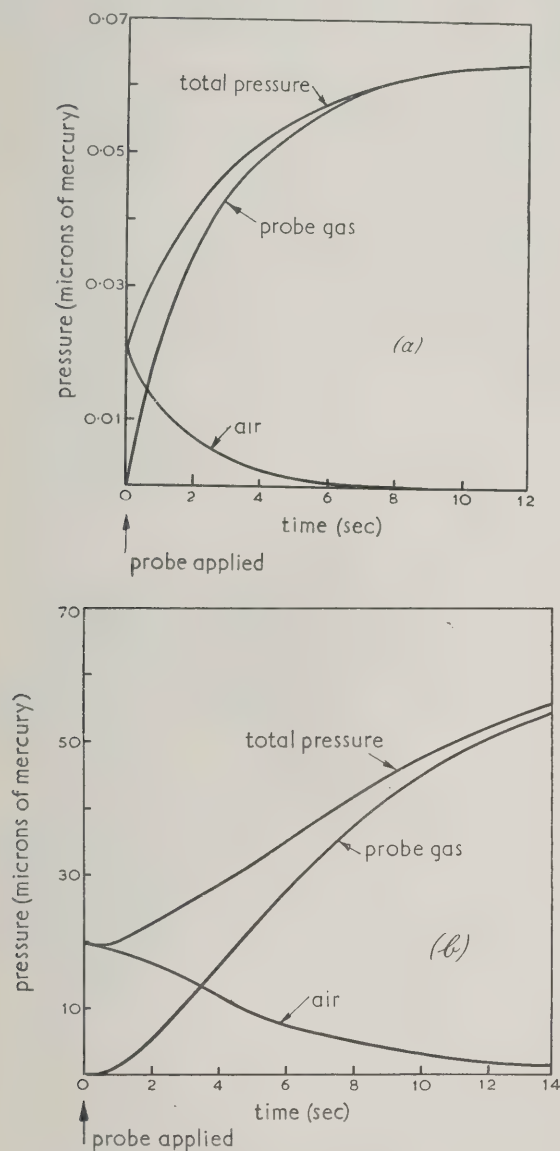
Fig. 3. Pressure changes after covering leak with Butane. (*a*) Fine side changes. (*b*) Backing side changes. Constants of system: $V' = 100\,1$; $V'' = 0\cdot2\,1$; $U' = 50\,1/sec$; $U'' = 0\cdot05\,1/sec$. Nominal time constants, fine side = 2 sec; backing side = 6 sec; leak rate = 1 $\mu l/sec$

3.2. *Transient pressure/time equations.*—The pressure/time relations of a leak detection system are determined by the time required for the probe gas to pass through the leak and displace the air from the vacuum system. The time lag of the gauge itself must also be

considered. For a single hole leak the delay in the leak is of very short duration ($3 \times 10^{-5}L^2/D$ sec is the average time spent by a molecule in diffusing through a hole of diameter D, length L). The response for the system shown in Fig. 2 can be calculated by neglecting the delay in the leak itself, and assuming that pumping of air and the probe gas take place independently.

If at the time $t = 0$ the leak is completely covered with probe gas the partial air pressures P_A', P_A'', will begin to fall, whilst the corresponding partial probe gas pressures will begin to rise. The pressure-time equation for the fine side is:—

$$P_A' + P_P' = \frac{L_A}{U_A'} \exp\left(-U_A't/V'\right) + \frac{L_P}{U_P'}\left[1 - \exp\left(-U_P't/V'\right)\right]$$
(9)

i.e. the pressure is the sum of a falling exponential for air and a rising exponential for the probe gas, the respective time constants being V'/U_A' and V'/U_P'.

For the backing side, conditions during the transient are much more complicated, because now two volumes and two contrictions are involved in the equations. A solution can again be obtained in terms of the rates at which air and the probe gas enter and leave the backing space, the pressure/time equation following complete coverage of the leak being:—

$$P_A'' + P_P'' = \frac{L_A}{V'U_A'' - V''U_A'}$$
$$\left[V'\exp\left(-U_A't/V'\right) - \frac{V''U_A'}{U_A''}\exp\left(-U_A''t/V''\right)\right]$$
$$+ \frac{L_P}{V'U_P'' - V''U_P'}\left\{\frac{V'U_P'' - V''U_F'}{U_P''}\right.$$
$$\left. - \left[V'\exp\left(-U_P't/V'\right) - \frac{V''U_F'}{U_P''}\exp\left(-Ut/V_P''\right)\right]\right\}$$ (10)

A more detailed treatment of the transient flow equations has been published by A. H. Turnbull.[18] Equations (9) and (10) can be used to plot response curves following the application of the probe, and typical curves for the fine and backing side of the plant shown in Fig. 2 are given in Figs. 3*a* and 3*b*. These results show that though the mean pressures on the fine and backing sides are very different, the fractional changes in total pressure are approximately the same in both cases. For the example chosen the pressure has been increased 1 000 times at the expense of increasing the time constant by a factor of 3. Furthermore, as Fig. 4 shows, the background pressure fluctuations for both fine and backing sides would be much smaller than the corresponding pressure changes resulting from probing. Figs. 3 and 4 therefore emphasize the fact that by using a constriction on the backing side to build up the pressure to 10 or 20 μ, the relatively insensitive Pirani gauge may be used almost as effectively as a fine side ionization gauge. It has been shown elsewhere[19], [21] that the optimum value of the time constants ($V'/U' + V''/U''$) is 10 sec. Apart from this, the requirements for

high sensitivity are exactly the same as those for obtaining maximum steady state change [e.g. equation (8a)].

3.3. *Pressure fluctuations.*—Instabilities in the operation of diffusion pumps give rise to pressure fluctuations with a period between 1 and 10 sec and an amplitude of about 2% of the mean gas pressure. The effect is observed on both the fine and backing sides of the pump. The gauges themselves also have inherent fluctuations even when no gas is being pumped. These two effects combine to limit the ratio of mean pressure/minimum detectable change of pressure (i.e. the ratio total leak/minimum detectable leak). For Pirani gauges and ionization gauges the situation is summarized by Fig. 4.

To minimize the effect of fluctuations the constriction may be given a value which reduces the speed of the
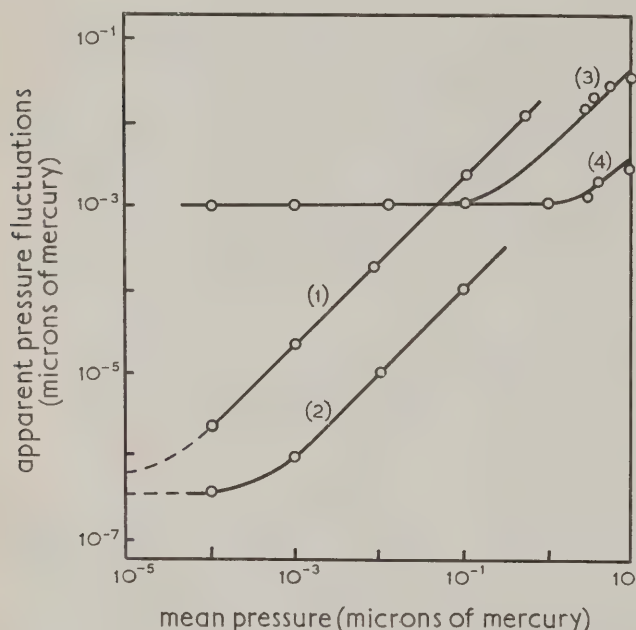
Fig. 4. Fluctuations in Pirani and ionization gauge readings as a function of gas pressure. Ionization gauges on fine side, Pirani gauges on backing side of 3 in oil diffusion pump. (1) Single ionization gauge, (2) differential ionization gauge, (3) single Pirani gauge, (4) differential Pirani gauge

diffusion pump by a factor of 10. The percentage fluctuation will also be reduced by factor 10 because the fluctuations, which originate below the constriction, remain constant in magnitude, whereas the pressure at the measuring point above the constriction is increased 10-fold. For fine side methods there is thus a good case to be made out for using large pumps, a typical combination being a 4 in pump (with a speed 200 l/sec) and a 20 l/sec constriction. Using an ionization gauge on the fine side this combination would be suitable for vessels up to 200 l in volume, and the fine side gas pressure fluctuations would not exceed 0·2%.

An alternative to constricting the pumps is to constrict the gauge tubulation until the gauge time constant is

several times the period of the fluctuations. The only objection to this expedient is that it slows down the degassing of the gauge, and thus increases the time required to obtain high sensitivity.

More elaborate methods for reducing the effect of pressure fluctuations are described below.

3.3.1. *The differential method.*—By attaching identical gauges to the same point in the vacuum system and connecting these in a bridge circuit it is clearly possible to balance out both the mean pressure and the pressure fluctuations. By preventing the probe gas from entering one gauge, whilst leaving it free to enter the other, a differential change of pressure will be observed during probing. This principle was first put to use by Jacobs and Zuhr[1] who used condensation of a hydrocarbon probe gas by means of a liquid air trap to produce the differential effect. A similar result has been achieved by using chemical absorption of carbon dioxide in a calcium hydroxide trap.[19] The latter method is not so sensitive, but it eliminates the costly liquid air and the gaseous hydrocarbon probe with its attendant fire risks. Ionization gauges or Pirani gauges may be used for detecting the differential change and it is found that, other things being equal, the differential connexion reduces fluctuations by factor of 20. This subject has been discussed in some detail in a recent paper by the authors.[20]

3.3.2. *The hydrogen–palladium tube.*—An ionization gauge with a palladium interface between the gauge and the vacuum system can be used to detect hydrogen. The palladium, when heated to 700–800° C allows hydrogen to diffuse freely whilst remaining impervious to other gases.[21], [22] Since the gauge is not affected by the air pressure or the fluctuations of air pressure, it can be attached to the backing space thereby giving the advantage of increased probe gas pressure without the loss of sensitivity imposed by using the Pirani gauge. It is unfortunate that a gauge with such apparent advantages is not well described in the literature. There are some practical difficulties due to spurious readings caused by the fact that oxygen absorbed on the palladium may restrict the diffusion of hydrogen and, so far as can be judged from the scanty evidence available, these difficulties make the palladium tube gauge rather less sensitive and reliable than the normal fine side ionization gauge.*

3.3.3. *The mass spectrometer.*—A particular advantage of the mass spectrometer is that it responds solely to the probe gas: consequently fluctuations of air pressure are without significance and the air pressure itself is only important when it causes scatter in the tube or endangers the filament life.

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* A. S. D. Barrett[22] informs us that by admitting a controlled amount of H_2 or O_2 to the vacuum system it is possible to maintain the correct stiochiometric relationship between oxygen and hydrogen and thus overcome this difficulty. A single adjustment of the "make-up" gas flow rate usually suffices for each test vessel, and leak rates down to 10^{-3} μl/sec may be detected easily. With small vessels (1 litre) and mercury pumped systems, a minimum detectable leak rate of 10^{-5} μl/sec has been achieved.

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3.4. *Probe gas.*—It is useful to compare the various probe gases in common use. Setting aside the considerations of fire risk, expense, danger to health, etc., the important point is the gauge deflexion on covering a given leak. As equation (8) shows, this deflexion is governed by the following effects: (i) the partial pressure of the probe gas in the atmosphere over the leak; (ii) the change in flow rate through the leak when a probe gas is substituted for air; (iii) the change in flow rate through the diffusion pumps when the probe gas is substituted for air; (iv) the relative sensitivities of the gauge for air and the probe gas; (v) in the special case of the differential gauge, the effectiveness of condensation or absorption.

The overall result of these effects has been measured by opening a needle valve to produce a definite air pressure in the gauge, then covering the leak with the probe gas and noting the apparent change in pressure. The ratio (change of pressure caused by covering leak with probe gas)/(mean air pressure before probing) is called the substitution sensitivity ϕ. The results for various probe gases and detector elements are shown in Table 2.

Butane–C_4H_{10} ("Calor gas") has been found to be the most satisfactory probe gas for both the ionization and Pirani gauges. Furthermore, since butane can be sprayed in the gaseous form there is no danger of temporarily blocking the hole as there is when using liquid probes such as benzine or carbon tetrachloride.

3.5. *Minimum detectable leak.*—The significance of ϕ is that when the fluctuations of air pressure ΔP_A at the gauge are known (Fig. 4) the minimum detectable leak can be found from the simple relation:—

$$\Delta L = \Delta P_A U_A / \phi$$

Table 3 lists values of ΔP_A and ϕ obtained experimentally, in a plant similar to that shown in Fig. 2. It may be useful to illustrate the use of this table by an example. Suppose the pressure at the gauge is $0 \cdot 1 \mu$, the conductance $= 5$ l/sec and the available instrument is a single ionization gauge. Then choosing butane as a probe we have:—

$$\Delta L = 5 \times 2 . 10^{-3} \times 1/10 = 10^{-3} \ \mu l/sec$$

or, more simply, since $L/\Delta L = 500$ and $L = 5 \times 10^{-1}$,

$$\Delta L = 5 \times 10^{-1}/500 = 10^{-3} \ \mu l/sec$$

Table 3 may be said to summarize the case for the simpler methods of leak detection.

4. PRACTICAL CONSIDERATIONS

4.1. *Virtual leaks.*—All vacuum systems contain relatively large amounts of surface adsorbate which is given off from the walls during pumping, thus giving rise

Table 2. *Substitution sensitivity factors (ϕ)*

Probe gas	ionization gauge single	differential	Pirani gauge single	differential
Butane	10	11	1	2
Di-ethyl ether	5	6	0·7	1·7
Carbon dioxide	1	2	0·3	1·3
Carbon tetrachloride	1	2	0·05	1·0
Benzine	0·3	1·3	0·1	1·1
Hydrogen	−0·4	0	0·4	0
Coal gas	0·25	0	0·25	0

Table 3. *Sensitivities of various leak detection methods*

Mean air pressure at gauge μ		Minimum detectable change in air pressure ΔP_A					Ratio of undetected to detectable leak $L/\Delta L$				
		0·001	0·01	0·1	1	10	0·001	0·01	0·1	1	10
Single Pirani with	hydrogen (2)						0·6	6	60	140	140
	hydrogen (1)						0·4	4	40	100	100
	carbon dioxide (2)						0·7	7	70	180	180
	butane (2)	0·001	0·001	0·001	0·004	0·04	0·5	5	50	125	125
	butane (1)						1	10	100	250	250
	acetone						0·2	2	16	40	40
Differential Pirani with	carbon dioxide (1)	0·001	0·001	0·001	0·001	0·005	1	10	100	1 000	2 000
	butane (1)						2	20	200	2 000	4 000
Single ion gauge with	hydrogen (2)	2×10^{-5}	2×10^{-4}	2×10^{-3}	—	—	8	8	8	—	—
	carbon dioxide (2)						50	50	50	—	—
	butane (1)						500	500	500	—	—
Differential ion gauge with	carbon dioxide (1)	10^{-6}	10^{-5}	10^{-4}	—	—	2 000	2 000	2 000	—	—
	butane (1)						10 000	10 000	10 000	—	—
Penning and Nienhuis gauge, gauges depending on work function		Approximately the same as single ionization gauge.									
Mass spectrometer with	helium	10^{-8}	Changes of *probe gas* pressure				2×10^4	2×10^5	—	—	—
	hydrogen	10^{-7}					2×10^3	2×10^4	—	—	—

(1) Authors' results. (2) B. A. Collett's[18] results.

Note.—Gauge deflexion on covering leak with the probe gas is ϕ times ΔP_A.

to gauge readings which may be mistaken for real leaks. If no cold trap is present removal of the adsorbate (degassing) produces an apparent or virtual leak of about 10 μl/sec per square metre after 15 min pumping, falling to 1 μl/sec after one hour and subsequently by approximately a factor of 2 per hour. This degassing introduces two difficulties: (*a*) it often leads to much loss of time in searching for non-existent leaks, (*b*) it increases the mean pressure in the vacuum system and thus lowers the sensitivity of the leak detector. A mass spectrometer capable of giving a measurement of the oxygen partial pressure is the only known way of distinguishing unambiguously between real and virtual leaks. Recent work[16] has shown that for well cleaned vessels about 75% of the desorbate is water. Consequently, by placing a water absorbing trap (such as phosphorous pentoxide) between the vacuum system and the gauge, a considerable improvement in performance can be obtained.

By covering the vessel under test with some form of close-fitting box or "hood" and filling this with the probe gas, the presence of the leak may be verified quickly (to limits imposed by the value of $L/\Delta L$, where L is the total leak, real plus virtual). Virtual leaks undoubtedly constitute one of the most serious disadvantages of the simple methods, since they force the operator to wait until the pressure drops sufficiently for him to obtain sufficient sensitivity.

4.2. *By-pass methods.*—It is common experience that large leaks ($P_A > 100 \mu$) are often more difficult to detect than the smaller ones, simply because they give rise to pressures outside the range of the leak detector. A plant such as that illustrated in Fig. 5 allows the major

part of the leak to be removed by a large pumping plant whilst a small fraction may be by-passed to the leak detector by means of a butterfly valve. When the large leaks have been closed a greater fraction of the leakage gas can be by-passed to the detector, thus increasing the sensitivity.

4.3. *Leaks in series.*—Two holes in series with a volume in between make a leak which is difficult to locate. (The condition sometimes arises when one weld is put down over another.) Suppose the smaller hole on the inside allows 0·1 μl/sec to pass into the vessel and that the volume between the two leaks is 0·1 c.c. then the time constant of the leak is of the order 20 min. Probing at normal speeds, where a 5–10 sec time constant is assumed, would thus result in a 100-fold loss of sensitivity. Any form of construction which is likely to give this condition (e.g. double welds, double gaskets) should therefore be avoided unless provision is made for separately pumping the dead space.

5. CONCLUSIONS

Before attempting to detect a leak in an *existing* plant the following steps are considered essential. (1) To establish that the leak is real. In the absence of a mass spectrometer the surest sign of a real leak is that the pressure remains constant at a high value or is not falling at a greater rate than indicated in Section 4.1. If the leak is very small the hood technique with complete coverage is of help. (2) To calculate the pumping speed, pressure and pressure fluctuations at the gauge and then calculate whether the gauge available is likely to be able to detect the probable change in pressure due to probing. (In using Table 3 note that the air pressure change detectable by probing is less than the value tabulated. It is $\Delta P_A/\phi$.) (3) To find the time constant of the system and recognize the necessity for covering each part of the test vessel for a time at least equal to the time constant.

In designing a *new* system care should be taken to see that points for attaching leak detection gauges are available on both fine and backing sides. A fine side valve can usually serve as a variable constriction and this valve should be supplemented by a variable constriction on the backing side.

The chief conclusion to be drawn from this review is that by a careful design of the system standard vacuum gauges can be used to detect all the important leaks on continuously pumped equipment, whereas arbitrary choice of pumping speed and backing space volume, and omission of constrictions can easily reduce the sensitivity to a small percentage of the value shown in Table 3. Often time is wasted in a vain attempt to find a small leak with either a Pirani gauge or a Geissler tube when, because the diffusion pumps are not operating the time constant is 100 seconds or more, the operator not realizing that coverage of the leak for approximately 100 sec is required to obtain a reasonable response. It has been our experience that a large leak (200 μl/sec)
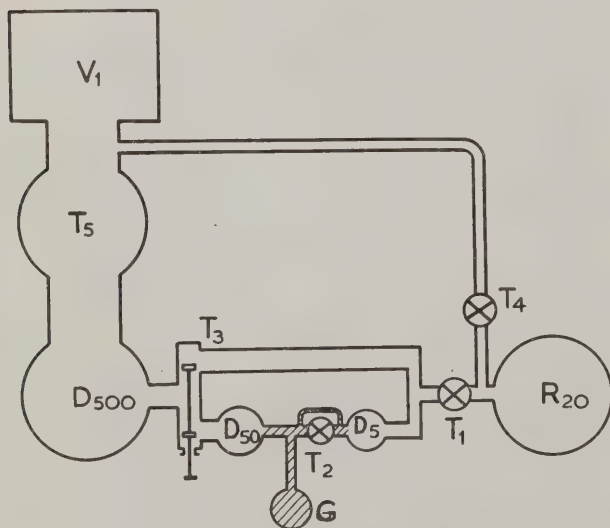
Fig. 5. Schematic arrangement of "By-pass" vacuum testing plant. V_1 test vessel; D_{500} D_{50} D_5, diffusion pumps; R_{20} rotary pump (subscripts indicate typical speeds) T_1 T_2 T_4 diaphragm valves; T_3 butterfly valve; T_5 fine side valve. The butterfly valve T_3 is used to select a measured fraction of the leakage gas, and T_2 is adjusted for optimum pressure at the gauge G

on a 100 litre vessel has been more difficult to locate than one of 0.01 μl/sec because with the large leak it has been impossible to run the diffusion pumps.

When stating the sensitivity of a new form of leak detector it has been the practice to give a value of the minimum detectable leak without reference to the design of the pumping system. This practice makes comparison of different methods very difficult and it would be better always to give the value of minimum detectable change of air pressure. With this figure it is possible to calculate the minimum detectable leak for any plant where the new method is applied.

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DISCUSSION

Mr. K. W. Brown: In the case of the differential ionization gauge leak detector, is there a difficulty due to the liquid air trap causing differential pumping for condensible gases present in the system, say water vapour?

Authors' reply: No practical difficulty is caused by the liquid air trap pumping water vapour, or other condensible vapours, after the system has been continuously pumped for a few (two to four) hours.

Mr. A. G. Hayes: What is the effect of the ionization gauge pumping the system during a leak measurement?

Authors' reply: This effect is of little importance except for very small leaks, but does actually effect an improvement. The so-called pumping by the gauge reduces the total pressure in the gauge without in any way reducing the signal when a leak is covered.

Dr. M. Pirani: The authors mentioned certain inconveniences in the degassing of ionization gauges, especially in the presence of oil vapours. Would they say a little more about these?

Authors' reply: For leak detection the degassing of the ionization gauge has been found to cause little practical difficulty. After the gauge has been running for a few hours the drift due to degassing is usually negligible. The only measure taken to speed up the degassing has been the use of a small electric heater to keep the glass bulb at a temperature of $100°$ to $150°$ C for two or three hours.

Dr. L. L. Katan: It was stated that carbon dioxide is the next best probe gas after butane. Does this refer to its use with a differential detector, or with a single Pirani gauge; and with a differential detector, is the elimination of carbon dioxide effected by calcium hydroxide or by liquid air cooling? Would not calcium oxide, which would not evolve water, be a more suitable absorbent than calcium hydroxide?

Authors' reply: In our experience carbon dioxide has been found to be the most suitable probe gas for the single Pirani gauge, after the inflammable vapours butane and ethyl ether. For the differential absorption method, carbon dioxide is the only probe gas used. However, it cannot be used for the differential method with a liquid air trap, since carbon dioxide is not

effectively condensed at liquid air temperatures. With calcium hydroxide the evolution of water vapour has not been found to be of practical importance. The presence of water vapour is noticed only when a large leak is covered for a long period, say five minutes. It has been found that calcium oxide does not absorb carbon dioxide and therefore the hydroxide must be used.

Mr. L. Breen: What are the objections to using liquids for probing?

Authors' reply: With rubber gaskets the rubber may swell, effectively closing the leak for a few hours. Further, a drop of liquid may seal the hole, thus resulting in an apparent fall in pressure instead of the expected rise. Normally, after the leak has been covered, it remains covered for some minutes so that a rapid check cannot be made.

Dr. L. Riddiford: We use liquid trichlorethylene probing for all our leak-hunting, with a single ionization gauge. Two advantages are that the leak can be pin-pointed accurately, and that large surfaces can be covered very rapidly. But the method is not a good one for leaks with a time constant of a few seconds, as for instance in the case of a narrow channel connecting two holes, one on each side of a double weld. Here all that happens is that the hole is blocked, and the pressure of probe vapour which appears in the system is much lower than with a short hole, because many of the molecules which evaporate from the liquid surface return to it. Consequently the amplification factor over air is about unity, instead of the normal twenty-five. Because of the large amplification factor, it is believed that the method would be a powerful one in a differential system, for leaks of short time constant. This factor arises firstly on account of the greater sensitivity of the ion gauge for the heavy vapour, and secondly because the liquid is forced into a short hole, so that the molecules evaporate directly from the surface into the system.

Mr. L. W. Owen: What type of jet is recommended when it is desirable to locate the position of the leak precisely?

Authors' reply: The actual position of a leak can be located by means of a very fine jet of gas. The flow of gas should be controlled so that it may be reduced as the exact location is determined more and more nearly.

Dr. F. Ashworth: The probe gas used could be localized successfully by means of appropriately shaped sheet rubber applicators into which the probe gas is introduced through rubber tubing.

Do the authors consider a differential Penning gauge to be a useful variant on the two types of leak-detection gauge discussed?

Authors' reply: The Penning gauge has not been studied for use in the differential method, but an investigation would be worth while. The single Penning gauge has been used in conjunction with "Calor gas," and has been found to be useful in a number of cases.

Mr. N. Ochert: Do the authors consider it is possible that some of the probe gas used in a previous test may become trapped and stored in some part of the system, or dissolved in, say, the rotary pump oil, and be released in a subsequent test, thus giving the impression of a leak?

Authors' reply: In practice the amount of probe gas which enters the system is small, and causes no difficulties in subsequent work.

Dr. D. A. Temple: The authors' suggestion for removing rust from surfaces to be subjected to a vacuum, i.e. by pickling, is open to comment. It is known that during acid-pickling there is absorption of the hydrogen which results from the solution of the metal, this being particularly true for steel, unless the pickling bath is ideally inhibited against metal solution. During subsequent vacuum treatment, this hydrogen might easily cause as much trouble by outgassing as the original rust. It would seem that either wire-brushing or shot-peening are preferable methods for removing rust or other corrosion products.

Authors' reply: We have experienced no difficulties due to the evolution of hydrogen brought about by pickling. In the light of mass spectrometer results the effect must certainly be one or two orders of magnitude less than the water vapour evolution.

Mr. I. Varnam: What is the sensitivity of the platinum diode leak detector, when used in compression testing, and is there any possibility of using this detector under vacuum searching externally with a halogen probe?

Dr. C. J. Milner: The sensitivity depends on the excess pressure. For one atmosphere the limit is about $0 \cdot 01$ to $0 \cdot 001$ μl/sec. There appear to be no advantages of using it under vacuum.

A cold-cathode mass spectrometer leak detector

By C. J. MILNER, M.A., Ph.D., F.Inst.P., Research Laboratory, The British Thomson-Houston Co., Ltd., Rugby

A cold-cathode spectrometer has the advantages of robustness, long life, and avoidance of refrigerated traps. In the instrument described, the use of an a.c. amplifier for the ion current, in conjunction with velocity modulation of the ion beam, greatly reduces background effects. Leaks smaller than 10^{-5} μl/sec can be detected in vessels whose gas output is up to 1 μl/sec. Gas outputs up to 100 μl/sec can be handled by the instrument's pumps, with proportional increases in detection limit. Various features of the design and characteristics are discussed. Some observations are made with regard to the applications and field of use of the instrument.

The method of detecting vacuum leaks with a mass spectrometer was developed in America during the war-time work on large-scale isotope separation.[1] In this method, a mass spectrometer is used as a "selective ionization gauge," responsive only to a particular gas. For this, helium is usually chosen, and the spectrometer is correspondingly set to mass 4. In practice, ions of this mass are produced only from helium, which is, practically, absent both from the gases liberated from the walls of a vacuum system and from the air which enters through leaks. Thus the spectrometer responds only when helium is applied to the external end of a leak through which it can enter the vacuum system. This response is obtained within a few seconds; the presence of a leak is therefore quickly detected. The application of helium through a suitable probe to various parts of the system in succession allows the location of invisible leaks within a few millimetres. The sensitivity of this method greatly exceeds that of more conventional techniques; using a mass spectrometer, leaks as small as 10^{-5} μl/sec or less can be detected. The leak detector described in this paper comprises a mass spectrometer together with its electromagnet, rotary and diffusion vacuum pumps, electrical supply and amplifier units and associated controls built into a trolley cabinet. It differs in important respects from
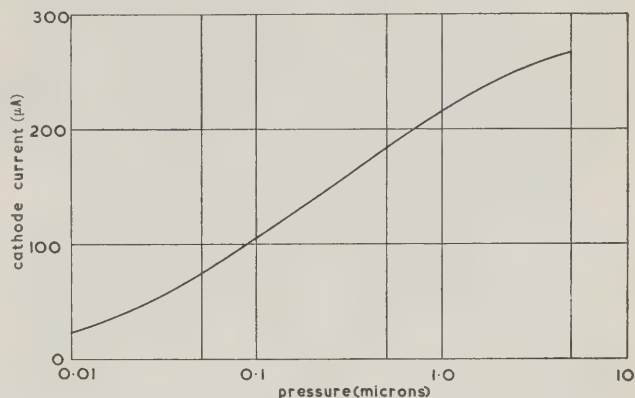
Fig. 2. Relation of cathode current and pressure. Air plus about 0.01 μ of residual gas. Valid for all magnet currents from 0.5 to 1.0 A (fields from 2 000 to 4 000 gauss)

most of the American instruments of this type,[2, 3, 4] although distinctly resembling one described[5, 6] since this paper was first prepared. The instrument comprises a mass spectrometer together with its electromagnet, rotary and diffusion vacuum pumps, electrical supply and amplifier units and associated controls, built into a trolley cabinet.

COLD-CATHODE ION SOURCE

The present instrument differs radically from the majority of mass spectrometers in employing a cold cathode in the ion source, in place of the usual hot filament, and the electron supply comes from secondary emission under positive-ion bombardment instead of thermionic emission. The hot cathode is reported, at least in leak detectors, to give only a life of a few hours, involving frequent replacement. This is probably due to the relatively high total pressure at which it must operate if a maximum sensitivity is to be secured. Even so, it appears necessary to use liquid-air or solid carbon dioxide traps in hot-cathode mass-spectrometer leak detectors to secure an adequate vacuum. The cold-cathode ion source avoids both of these objections. The cathode life before replacement is at least many hundreds of hours, and the optimum total pressure is in this case so high that refrigerated traps are unnecessary.

The electrode arrangement in the cold-cathode ion source is shown diagrammatically in Fig. 1. The electrode structure is immersed in the uniform magnetic
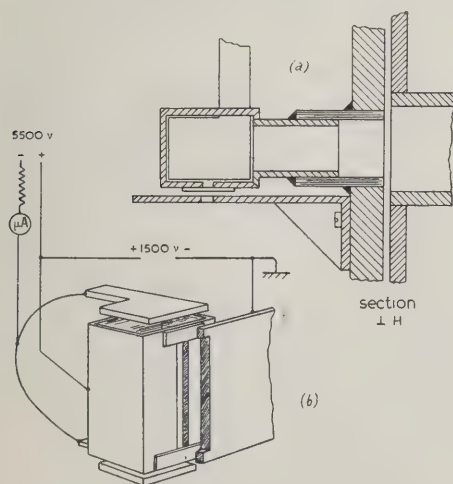
Fig. 1. Diagrams of cold-cathode ion source: (a) mid-section, perpendicular to magnetic field; (b) perspective view (part cut away), and circuit

field of the spectrometer: as in the Penning gauge, the cathode consists of a pair of parallel plates, spaced apart in the direction of the magnetic field, and a hollow anode is placed between them. Here, however, the anode is a metal tube, two or three diameters long, whereas in the Penning gauge a ring of wire is normally used. A slit in the tube wall parallel with the length, and a second slit, adjacent to the first, in a plate lying alongside the tube and at a negative potential with respect to the tube, constitute the ion beam gun. Data on ion sources of this type have been published by Thomas.[7]

Fig. 4. Block diagram showing components of mass spectrometer leak detector

In the present instrument, a high-tension supply feeds the cathodes, through a resistance of 20 MΩ, from a

Fig. 3. Vacuum system assembly

point — 5 500 V to the anode. The current flowing in the cathode lead may then be used, as that in a Penning gauge is used, as a convenient measure of the gas

pressure in the inter-electrode space. Fig. 2 shows a calibration curve for the arrangement employed. It is found that the optimum condition comes at a cathode current of about 200 μA, corresponding to a pressure of roughly 1 μ; one says "roughly" because, as commonly found in Penning gauges, the exact curve relating current and pressure has numerous kinks and discontinuities which vary a lot with conditions and from time to time. They do not, however, at any time deflect the relation farther from the smooth curve shown than about $\pm$ 20 μA, or a factor of two either way in pressure. At this optimum pressure, and with 1 500 V applied to the ion gun, the beam divergence is about $\pm$ 6–8°, and the ion energy spread is about $\pm$ 3–5%.

MASS SPECTROMETER AND VACUUM SYSTEM

The mass spectrometer in the present instrument is of the 180° type with 4·5 cm radius. The electrode system is all mounted on a removable faceplate, inserted into the rectangular metal vacuum chamber shown, together with the main vacuum system, in Fig. 3. This chamber lies between magnet poles which are U-shaped so as to direct the flux from the electromagnet core to the region of the ion beam where it is needed. The vacuum system is designed to dismantle into four units which can be easily removed from the trolley chassis and reassembled as shown. When in position the system is entirely enclosed (for high-voltage protection) and all operating controls are grouped on an external panel.

The block diagram, Fig. 4, shows the general arrangement and functioning of the vacuum system. The vessel to be tested is attached to the flange indicated at the top left.
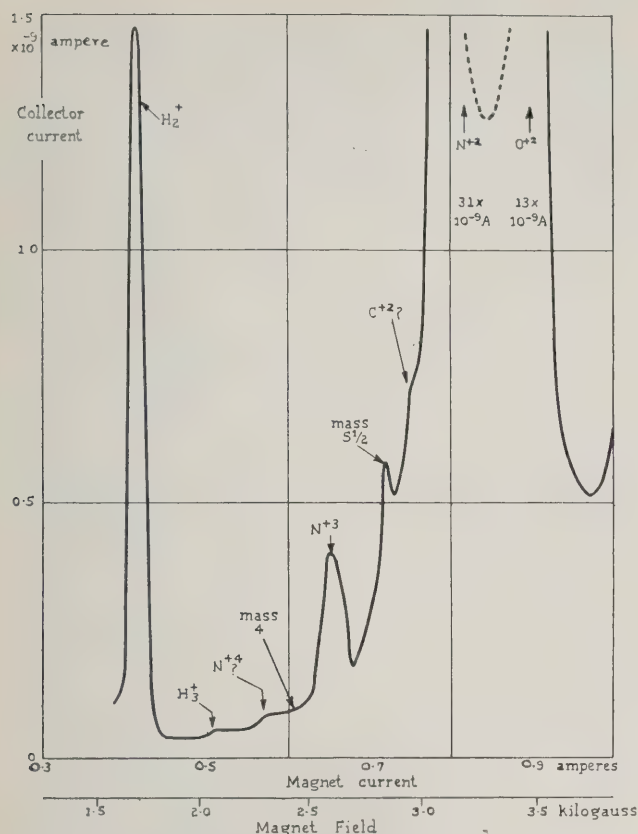
Fig. 5. Mass spectrum obtained with d.c. amplifier. Air at $0.8\,\mu$ plus about $0.01\,\mu$ of residual gas. (The superior index $+n$ indicates a positive ion carrying n electronic charges)

USE OF A.C. AMPLIFIER

A typical mass spectrum, obtained by varying the magnetic field at constant accelerating voltage, is shown in Fig. 5. This shows the collector current (as measured by a d.c. electrometer-valve amplifier) as a function of the magnet current and magnetic field, in the absence of helium from the system. It will be apparent that a considerable background exists in the neighbourhood of mass 4, against which helium has to be detected. Much of the undesirable background is eliminated in the instrument by the use of an a.c. amplifier, sharply tuned to 100 c/s, in combination with modulation of the ion beam velocity at 50 c/s, so that the helium line when it appears is swept back and forth across the collector slit. Such a line in the mass spectrum gives rise to 100 pulses of collector current per second. The fundamental component of this current is picked out and amplified, whereas the only effect of the velocity modulation on the background is to substitute one portion of the background spectrum for another. A uniform background, or even one having uniform slope, will produce no 100 c/s component of collector current; *curvature* of the intensity versus mass number curve is necessary to produce any response from this a.c. system. Thus a helium-ion current of less than 10^{-12} A will produce an

output indication as great as that from a background (see Fig. 5) of 10^{-10} A.

The system has the effect, which would be inconvenient to some mass spectroscopists, of making each line in the mass spectrum appear three times over. Let it be supposed that the ion beam voltage is 1 500 V d.c. plus 100 V 50 c/s ripple, as shown in the upper graph, Fig. 6; at a certain magnet setting, pulses of current due to a line in the mass spectrum will arrive at the collector when the voltage is at its mean value of 1 500 V, as in the middle one of the five lower figures. At certain slightly higher fields, pulses due to the line will be collected at, successively, 1 570 V and 1 600 V; and they will therefore be spaced in time as in the second and first of these figures respectively ($70 = 100 \sin 45°$, approx.). At certain lower fields the pulses will occur at 1 430 V and 1 400 V, and so the fourth and fifth figures will result. It will be seen that, in the first, third and fifth figures, the pulses are all spaced by a whole number of cycles of a 100 c/s wave, and therefore reinforce each other's effect on the tuned amplifier, whereas in the second and fourth figures the pulses occur in pairs whose members are a half-cycle apart and, therefore, cancel each other's effect. The resulting 100 c/s components of collector current are shown by broken lines in Fig. 6. If, as in this simple analysis, the focusing and resolution are perfect so that the pulses are of infinitesimal duration, the "side-lobe"
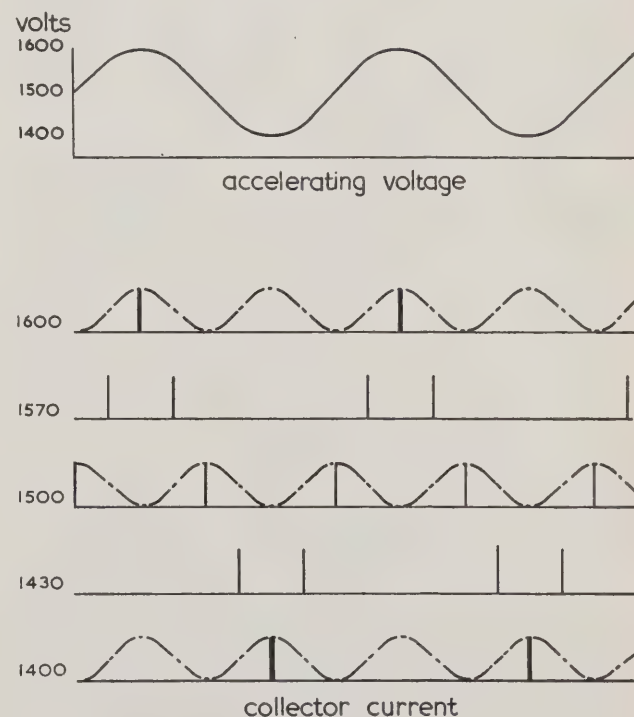
Fig. 6. Diagram showing origin of "triplets" in mass spectrum when a.c. amplifier is used. Broken lines show the 100 c/s component of collector current due to pulses arriving at the collector with time-spacings as shown

maxima are of equal amplitude with the centre-lobe (they are, incidentally, in anti-phase with it). In practice, however, the finite width and asymmetrical shape of the actual peaks in the spectrum make the triplet of output maxima due to any one mass unequal in amplitude, the lower side-lobe being about 40% and the upper about 80% of the central maximum, with a ripple setting of about ± 100 V, as used. The side-lobes due to mass 4 then appear at magnet settings corresponding to the centre-lobes which would be produced by masses of about $3\cdot7$ and $4\cdot3$.

OPTIMUM PRESSURE CONSIDERATIONS

For leak detection, the magnet current is set for the centre-lobe of the triplet due to helium. The cathode current in the ion source is then set to an optimum value by adjustment of the total gas pressure. The upper curve in Fig. 7 shows the amplifier output as a function of cathode current (varied by introducing air through the gas inlet valve, Fig. 4), when a constant partial pressure or throughput of helium is maintained from a separate test leak. Discontinuities such as that here recorded appear to correspond with prominent discontinuities in the cathode current/pressure curve which are, as previously remarked, rather transitory phenomena. The feature which should be noted from this curve is the pronounced maximum in the response at about 200–$210\ \mu$A. It will also be observed, from the lower curve, that the background response in the absence of helium, which is roughly constant at low pressures, increases rapidly at pressures above the optimum determined by the upper curve.

It cannot be said that these noteworthy phenomena are at all fully understood, though it is probable that both the increase of background and the decrease of signal (i.e. response to helium) observed at high pressures are due to scattering of the ion beams in the deflexion chamber. It is also probable that the loss of signal at low pressures is due to collection by the ion source cathodes of an excessive proportion of the ions formed. It may be noted that even under the optimum conditions the proportion of ions entering the beam is only small; only one ion reaches the collector per thousand or more molecules entering the ion source.

The origin of the observed background is also to some extent a matter for speculation. The nearest peak to the helium line identifiable in the d.c. mass spectrum of Fig. 5 is that due to N^{+3} at a mass number $4\frac{2}{3}$, and the lower side-lobe due to this peak seems to contribute a little to the background at mass 4, as also does the curvature of the tail of the large N^{+2} peak. Experimentally, however, trouble with background has been associated with the presence of residual organic vapours, and has been largely eliminated by the use of silicone oil instead of Apiezon oil in the diffusion pump. These vapours might, of course, give rise to the ions C^{+3} and perhaps O^{+4} which would be indistinguishable from

He^{+1}, but the observed background due to such vapours shows no special concentration at mass 4. It is suggested that possibly this diffuse background, which is particularly noticeable between mass numbers about 4 and 6, is due to numerous cases of dissociating polyatomic ions.[8] It can be shown that ions, having
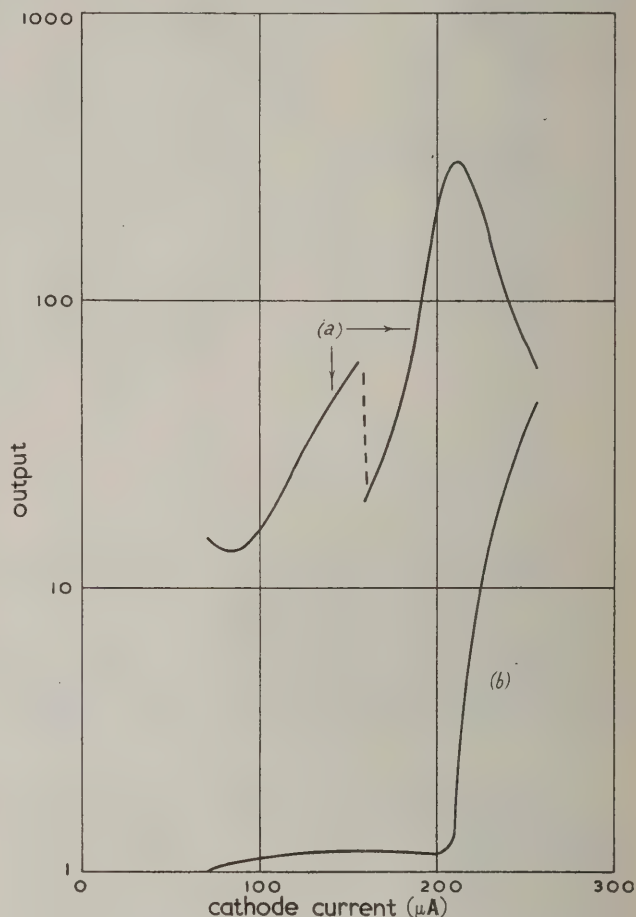
Fig. 7. Relation of output current and cathode current: (*a*) helium throughput $5 \times 10^{-4}\ \mu$l/sec; (*b*) no helium. Cathode current varied by admitting air

mass M_1 while accelerated, which then dissociate before deflexion so that the charge is carried by a fragment of mass M_2, will appear in the spectrum at the position normally associated with ions of mass M_2/M_1. With various complex ions (masses M_1), each of which can split into various fragments (masses M_2), the number of such possibilities is quite large, and the spurious lines which they set up would be too close for resolution with the present instrument—particularly when each "line" is a triplet!

OTHER DESIGN PARAMETERS

While discussing optimum conditions, it may be useful to comment on the compromise values assigned in the design to certain parameters.

Beam voltage and field. The accelerating voltage was fixed at 1 500 V because with this type of ion source the ion beam current at constant partial pressure increases rapidly with voltage, and the highest value consistent with reasonable size of magnet was therefore chosen. (The magnet weighs about 50 kg, and requires about 50 W for normal excitation.) The necessary field and voltage are related with the curvature of the ion path by the equation

$$MV = 48B^2R^2$$

where M = mass number focused,
V = beam energy (volts),
B = magnetic field (kilogauss),
R = radius of ion path (cm).

The radius of 4·5 cm was chosen for convenience; changes in this, keeping BR constant, do not markedly affect the magnet requirements. The magnet gap, which does strongly affect the magnet size, was fixed at 32 mm as the smallest value which would accommodate the spectrometer chamber walls, the cathode plates, and an anode of reasonable size, together with necessary clearances.

Cathode supply circuit. The cathode supply voltage was fixed at 5 500 V as the highest value practicable, so as to make the cathode current/pressure curve a sloping one over as wide a pressure range as possible. With this voltage fixed, the series resistance was fixed at 20 MΩ on the basis that lower values were found to give reduced sensitivity, and higher values to make adjustment to optimum pressure too critical. (The optimum pressure rises slightly as the series resistance is reduced.)

Pumping speeds. The throughput under optimum conditions is equal to the optimum ion source pressure multiplied by the pumping speed effective at this point; and it is desirable that this throughput should be as small as possible. The pumping speed at this point is measured to be 1·5 l/sec, and cannot easily be made less owing to the need for slits, and clearances for electrical insulation, having reasonable dimensions. The pumping speed at the spectrometer chamber should, on the other hand, be as high as practicable, to minimize scattering during deflexion; a value of approximately 30 l/sec is secured by the use of a rectangular pipe ($2\frac{1}{2}$ in × 1 in waveguide) giving a wide connexion to the chamber.

PERFORMANCE

The optimum total pressure in the ion source for operation of the present instrument is seen from Figs. 7 and 2 to lie at about 0·8 μ. Combining this figure with the speed of pumping measured at the ion source (1·5 l/sec), it is seen that there is, alternatively, an optimum total gas throughput of just over 1 μl/sec. Under this optimum condition, the curves of Fig. 7

show that a signal-to-background ratio of 250 is obtained for a helium throughput of 5×10^{-4} μl/sec. Hence it may be inferred that under these conditions a helium influx of 2×10^{-6} μl/sec would give a signal-to-background ratio of unity and so would be just detectable.

The "signal" shown in Fig. 7 was found to increase to half its final value in a time of 2·5 sec approximately on suddenly applying helium to the leak.

AUXILIARIES

Before proceeding to discuss the application of the instrument in leak detection, brief mention should be made of some of the auxiliary items included in the equipment, and indicated in the block diagram of Fig. 4.

Amplifier output indicators. The amplifier is provided with a panel-mounted output current meter, which, together with the gain control, gives a semi-quantitative indication of the size of leak which has been encountered. In addition the amplifier contains a circuit which develops a series of clicks from a telephone earpiece which serves as a portable output monitor. The frequency of clicks is arranged to rise from a preset value of about 1 or 2 per second to about 500 per second as the output current meter moves from zero to full scale.

Helium indicators. The rate of consumption of helium may be indicated by the passage of bubbles of this gas through a Wolff bottle built into the cabinet. In place of water in this bottle, a suitably dilute solution of ammonia may be used, and the helium thereby "loaded" with an odour which gives warning of undue escape of the gas.

When "bell jars" or other "hoods" are used to cover items suspected of leaking, they may be filled with helium by displacement, and a helium indicator, consisting of a simple form of thermal conductivity gauge, is provided, which can be inserted in the open bottom of such a jar to indicate when filling with helium is complete.

APPLICATIONS TO LEAK DETECTION

The adjustment for optimum total pressure depends on the conditions encountered. If the total gas output of the vessel under test is less than the 1 μl/sec throughput required at the ion source, air may be added through the gas inlet valve to make up the deficit. Alternatively, the throughput required to maintain optimum pressure may be reduced by partially closing the throttle valve, Fig. 4; this procedure allows an increase of sensitivity by a factor of 2 or 3 to be obtained with a proportional increase of response time. If, on the other hand, the total gas output exceeds 1 μl/sec, optimum conditions can be secured by opening the by-pass valve and partially closing the mass spectrometer inlet, so that

only sufficient gas to give the optimum throughput passes to the spectrometer, the surplus being withdrawn direct to the rotary pump. In this way, gas outputs up to about 100 μl/sec can be handled, the limit coming when the rotary pump can no longer maintain the backing pressure required by the diffusion pump. Of course, in this arrangement, sensitivity is lost proportionately as the fraction of total gas passed through the spectrometer falls.

Vessels or systems having a total gas output exceeding 100 μl/sec require auxiliary pumps. Fig. 8 illustrates
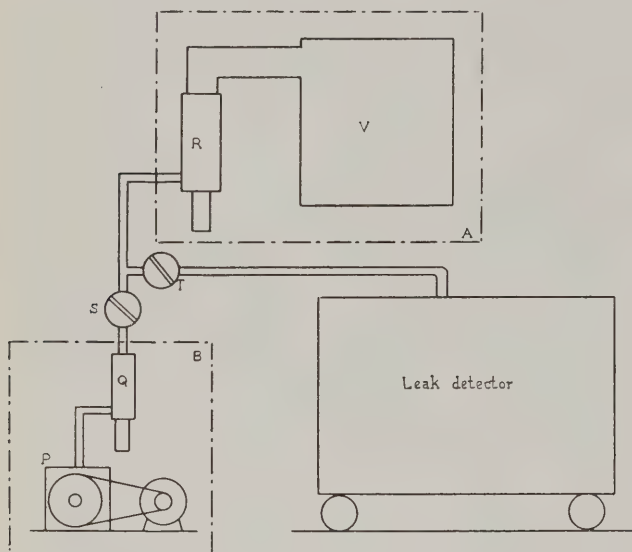
Fig. 8. Diagram showing uses of external pumps

some suggested arrangements. First consider box *A* simply as such a "gassy" system. A large rotary pump *P* may be used to withdraw most of the gas, and a fraction only, controlled by tap *T*, may be drawn off to supply the leak detector with the amount which it can handle. The diffusion pump *Q* shown in box *B* may sometimes be used with advantage, essentially to function as a non-return valve. Often, a rotary pump exhausts a system efficiently of its contents (which in this case are helium-bearing) and replaces them with (non-helium-bearing) oil vapours and/or atmospheric air. When the pump *Q* is in action, however, it prevents the "replacement" gas reaching *T*.

Turning to another aspect, if the vessel *V* is of large volume, a high-speed pump such as *R* may be used to sweep out the contents of *V* rapidly, and deliver them to the leak detector, with advantage in reduction of the response time. If in this case the total gas output is not excessive, tap *S* may be closed and the pumps in box *B* are not required.

CONCLUSION

A prototype of the instrument described has been in use in the author's laboratory for over two years, and has been successfully applied to a variety of leak detec-

tion problems. It can locate leaks so small as to be untraceable by any other method; other leaks have with its help been detected and located in a very small fraction of the time otherwise required. Its contribution to the improvement of vacuum technique is therefore clearly of great value.

As an illustration, one particular case may be mentioned. In the recent development of a specific type of u.h.f. valves, much trouble was early encountered with valves going soft in a few days, on standing after being sealed off. A considerable number of such valves were tested on the mass spectrometer, and in approximately three-quarters of the cases a leak was detected; in two-thirds of these it was also located to a specific part of the valve. In consequence of this work, efforts to improve constructional technique could be concentrated one one or two specific details, and the troubles were fully overcome.

It is of considerable interest to note that in this work many leaks of all sizes down to the smallest detectable were in fact detected, that is, that leaks as small as 10^{-5} μl/sec do exist and persist. (Very small leaks often shut themselves, at least temporarily.) The fact that such do exist is noteworthy new evidence in favour of pursuing still farther improved methods of leak detection, particularly when considered together with the importance in valve engineering of preventing softness, such as would be occasioned by a leak of even 10^{-7} to 10^{-9} μl/sec, if persisting over years.

It should be recognized, however, that increases of sensitivity would be of essential value only to the engineer of sealed-off devices; the present sensitivity of the mass spectrometer method is amply adequate for detecting all leaks that can give trouble in any conventional form of continuously pumped apparatus.

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DISCUSSION

Mr. A. M. Voyce: Would Dr. Milner define the method of measuring the signal-to-noise ratio in the mass spectrometer leak detector? The matter is of importance in comparing one type of instrument with another.

Author's reply: At present, comparison of leak detector systems is made by reference only to the order of magnitude of detection limits, and to attempt even one-figure accuracy is rather pedantic. Specifically, however, the "background," against which a helium "signal" is to be detected, consists partly of noise, and partly of coherent 100 c/s components from the mass spectrum, magnet ripple, hum, etc., whose resultant has undefined phase relationship with the helium signal. It is arbitrarily assumed that a signal will be detectable if its r.m.s. value is equal to the r.m.s. background, i.e. that on appearance of such a signal r.m.s. output will detectably increase.

Mr. J. Grindley: Dr. Milner has referred to the time of response of the leak detector. It should be made clear that this time is dependent also on the geometry of the vacuum system under test, and that the nature of the gas flow through the equipment needs study so that the detector may be used to best advantage. Leak-hunting becomes exceptionally tedious if the time of gas-flow from the leak to the source is greater than, say, five to ten seconds.

Mr. N. Ochert: Will the author describe the leak used by him in the measurements referred to in his paper, and give his opinion as to whether such a leak is of the same type (apart from steady-state throughput) as those expected to be encountered in industrial leak testing? A long hole and a porous patch may have the same throughput, and yet show a very different response when tested by such dynamic leak test methods as are in common use.

Author's reply: Speed of response is of great importance, because leak-testing needs to be carried out as rapidly as possible. The response time is controlled by several factors, including: (*a*) the time required for helium to flow through the instrument, (*b*) that required for the leak to fill the vessel under test to its equilibrium pressure. These factors are briefly discussed in the paper. There is also (*c*) the time required for helium to pass through the leak. The test leak used was a piece of copper tube, approximately $\frac{3}{16}$ in outside diameter and $\frac{1}{16}$ in inside diameter, hammered flat until the leak was of a suitable size, as measured by inflow of air to a McLeod gauge. It thus probably consisted of one passage, or two in parallel, roughly circular in cross-section, each being

about 2 cm long, and was therefore not untypical of many "airline" leaks.

Leaks of the same steady-state throughput may differ in response time, and I have encountered examples ranging from one second to one minute or more. Long response times may be due to cavities joined to the exterior by one leak, and to the interior by another, or to porosity. A single uniform circular hole, of length L cm and diameter D cm, has a pumping speed of about $10D^3/L$ l/sec under conditions of molecular flow; since in leaks of the size under consideration here, such conditions hold even up to atmospheric pressure, the leak rate

$$R = 7 \cdot 6 \times 10^6 \ D^3/L \ \mu\text{l/sec}$$

An approximate value for the response time for the hole is obtained by dividing its volume by its pumping speed, so that

$$T_{\text{hole}} = 0 \cdot 015 \ L^{5/3} R^{-\frac{1}{3}} \text{ sec}$$

Thus for $L = 1$ cm, if $R = 10^{-5}$ μl/sec, $T_{\text{hole}} = 0 \cdot 7$ sec; if $R = 10^{-8}$ μl/sec, $T_{\text{hole}} = 7$ sec. Now consider a porous area containing N such holes in parallel, each of leak-rate R. If the leak-rate for the whole area is F, the response time

$$T_{\text{porous}} = 0 \cdot 015 \ L^{5/3} F^{-\frac{1}{3}} N^{\frac{1}{3}} \text{sec}$$

We may suppose that an area of 1 cm² is involved, leaking at a rate $F = 10^{-5}$ μl/sec. N may range up to 10^{15}, if every path through the crystal lattice constitutes a leak, and so T_{porous} may range up to $0 \cdot 7 \times 10^5$ sec, or about 20 hours! It may well be that some "untraceable leaks" are due to porosity of this type. The term "porosity" is usually associated, however, with a countable number of individual metallurgical defects. Even 1 000 such defects contributing equally would raise the response time to only 7 sec. In my opinion, if a leak known to have been present cannot be traced with a leak detector of adequate (steady-state) sensitivity, it is more likely to be because the leak has stopped than because excessive response time prevents its detection.

Mr. J. Blears: Dr. Milner has shown that, when the total leak is *large*, the cold-cathode mass spectrometer may be at least as sensitive as, and possibly a little more sensitive than, the more common hot filament instrument. However, a fundamental limitation on the performance of cold-cathode instruments is imposed by the fact that ions are not produced effectively unless the pressure in the ionization chamber is about 10^{-4} mm

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of mercury. This means that partial pressures of helium less than 10^{-10} to 10^{-9} mm cannot be detected. No such limit is imposed on the hot-filament instrument, which can usually be relied upon to detect a partial pressure as small as 10^{-11} mm. Thus, for very small leak-rates, the hot-filament instrument is between ten and one hundred times more sensitive, and for this reason it will be preferred for many of the more exacting leak detection problems.

Moreover, ions formed by bombardment with thermionic electrons have small initial energies. Therefore an instrument with a hot filament may have a resolving power about one order of magnitude greater than one in which the ions are drawn from a gas discharge. Advantage can be taken of this fact in two ways: either the ion accelerating voltage can be reduced tenfold, with a corresponding reduction in magnetic field, or the high voltage can be retained with a correspondingly high resolving power. In the former case a very small

spectrometer tube and magnet, weighing as little as 2 lb, can be constructed; in the latter case heavier gases (such as butane) can be used instead of helium as the probe gas. From the practical point of view these are considerable advantages.

Author's reply: Mr. Blears indicates correctly that the cold-cathode mass spectrometer leak detector is especially useful where the gas output of the leaky vessel is large, i.e. above about 10^{-2} μl/sec, and emphasizes the advantage in detection limit of the hot-cathode spectrometer when the total leak-rate is below 10^{-3} μl/sec. In the author's experience, whether the vessel under test has been previous "degassed" or not, it takes at least an hour (sometimes much more) for its gas output to fall below 10^{-2} μl/sec. Thus a system which has superior performance only below 10^{-3} μl/sec can only be effective after a delay which destroys the greatest advantage of a powerful method of leak detection, viz. that it speeds up leak testing.

Application of the mass spectrometer to high vacuum problems

By J. BLEARS, B.Sc., A.M.I.E.E., Metropolitan-Vickers Electrical Co. Ltd., Trafford Park, Manchester

The potentialities of the mass spectrometer in studies of high vacuum problems are illustrated by descriptions of analyses of residual gases during initial evacuation, during baking and at the ultimate pressure. Surface exchange and synthesis products are considered, and it is concluded that mass spectrometry can solve many problems where it is difficult to distinguish between extraneous effects and real changes of pressure, or gas composition.

The mass spectrometer may be applied to high vacuum problems in three ways: first as a leak detector[1]; second for analysing the gases evolved in the processing of constructional materials, and in the operation of high vacuum electronic valves[2]; third for analysing the residual gases in normal dynamic vacuum systems. This paper is concerned with the latter aspect. The mass spectrometer itself, designed for analysing gases by measuring the relative quantities of ions produced from the different molecules present, is a typical small high vacuum system, and from observations of the residual gases in it under various conditions of operation it is possible to infer what gases may be present in other systems under similar circumstances.

The experimental results presented here were obtained with two types of mass spectrometer having quite different forms of construction. One was made from borosilicate glass and copper and was entirely free from demountable joints. This tube used mercury diffusion pumps and was basically similar to those described by Nier[3] and by Graham Harkness and Thode.[4] It was a typical example of the construction used in "very high" vacuum apparatus.[5] As examples of the more con-

ventional "dynamic" vacuum system two further tubes were used; one of these was made principally of brass, the other of stainless steel. In both of these tubes rubber gaskets were used freely and oil diffusion pumps produced the main vacuum.[6] Conventional ionization gauges were mounted on side arms attached to the cold traps of all the instruments used. Volumes, surface areas and other relevant constants are given in Table 1.

Table 1. *Data concerning the two mass spectrometers*

Construction	"Very high" vacuum (V.H.V.)	"Dynamic" (D)
Volume	3 l	4·5 l
Surface area	2 500 cm^2	4 000 cm^2
Pump speed (at tube)	5 l/sec	20 l/sec
Pump fluid	Mercury	Various oils
Constructional materials	Pyrex Copper	Brass Stainless steel
Gaskets	Nil	60 cm × 3 mm dia. pure rubber
Cold trap temperatures	− 78° C − 170° C	20° C − 78° C

The mass spectrum of residual gases is relatively complex. Hydrogen, water and oxygen give unique lines, but carbon monoxide and dioxide can only be separated from hydrocarbon by calibration and subsequent analysis. Fig. 1 shows part of the residual spectrum for the brass mass spectrometer tube using Apiezon C oil in the diffusion pump. Without a cold trap the hydrocarbon
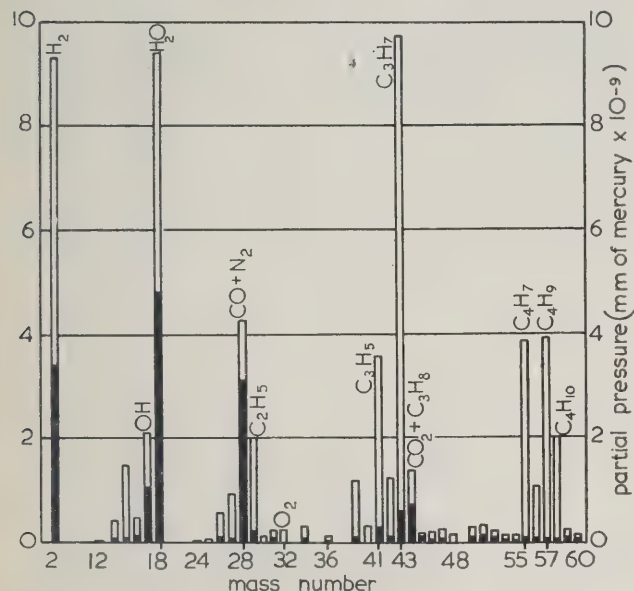
Fig. 1. Mass spectrum of residual gas in demountable mass spectrometer. Black part of line shows partial pressure with trap temperature $-78°$ C. White part shows the increase when trap temperature was raised to $20°$ C. (Oil diffusion pump.)

spectrum continues (beyond what is shown in the figure) up to mass numbers of several hundred. All spectra considered in this work have the *form* of Fig. 1, though the relative line intensities differ according to the experiment.

Some Typical Experiments

(1) initial evacuation

A stainless steel mass spectrometer tube was opened to the atmosphere for about 3 h and then evacuated,

the rotary and diffusion pumps being switched on simultaneously. Analyses of the residual gases, calculated from the mass spectra, are plotted in Fig. 2, from which some general conclusions may be drawn. For example, in the absence of leaks, it is comparatively easy to remove air from the tube. After 20 minutes the oxygen pressure was less than 10^{-7} mm of mercury: after 1 h it was 10^{-8} mm and still falling. Air, in fact, accounted for less than 0.5% of the material in the gas phase. About 70% of the ions present were formed from water, most of the remainder being from hydrocarbon vapours. Apart from the water, the largest single line in the mass spectrum was from carbon monoxide. This gas had a concentration of about 3% of that of the water.

The time constant for removal of residual molecules, deduced from the observed rate of fall of pressure, was only $1/4\,000$ of that calculated from the volume of the system and the speed of the diffusion pumps. Thus, though fast pumps can *maintain* a lower pressure than slow ones could, it is the forces binding molecules to the surfaces which determine the *rate* of removal of matter from the vacuum system. This conclusion could have been reached without the use of a mass spectrometer, but the surprising fact emerges that, at this early stage of an evacuation process, the concentrations of all kinds of molecule diminish at approximately the same rate.

If pumping (without baking or using cold traps) is continued for an extended period, the water content is gradually diminished, whereas the hydrocarbon (which
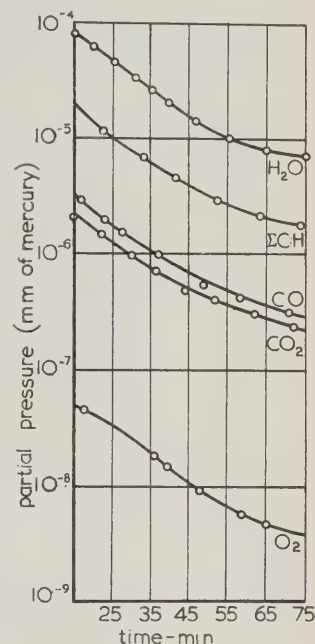
Fig. 2. Changes of partial pressure during initial evacuation of stainless steel demountable tube. (Oil diffusion pump.)

Table 2. *Gas analysis at the ultimate pressure*

All values quoted are after several weeks' operation with bakings for at least 48 h.   Units 10^{-9} mm of mercury

Pump fluid	Mercury			Apiezon BW		Apiezon C	
Baking temperature, °C.	100	250	100	100	100	100	100
Trap temperature, °C.	-78	-78	-170	-78	20	-78	20
	(1)	(2)	(3)	(4)	(5)	(6)	(7)
Hydrogen	—	—	—	3	22	4	9
Water	19	2·2	6·0	7	25	4	9
Carbon monoxide	9	0·1	1·0	1·1	—	3·1	2·5
Air	1·0	0·5	1·8	1·6	—	0·5	1·5
Carbon dioxide	1·3	0·1	1·7	0·5	12	0·9	1·8
Σ CH	4	1·2	1·3	5	1 250	10	60
Mercury	205	224	15	—	—	—	—

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comes from the pump fluid) does not fall below a limiting value. Corresponding analyses for a brass tube using two representative pump fluids are given in columns 5 and 7 of Table 2.

(2) ANALYSES OF RESIDUALS DURING BAKING

Fig. 3 illustrates the effect of low temperature baking. After an apparently constant residual pressure had been reached the system was baked at 100° C for an arbitrary
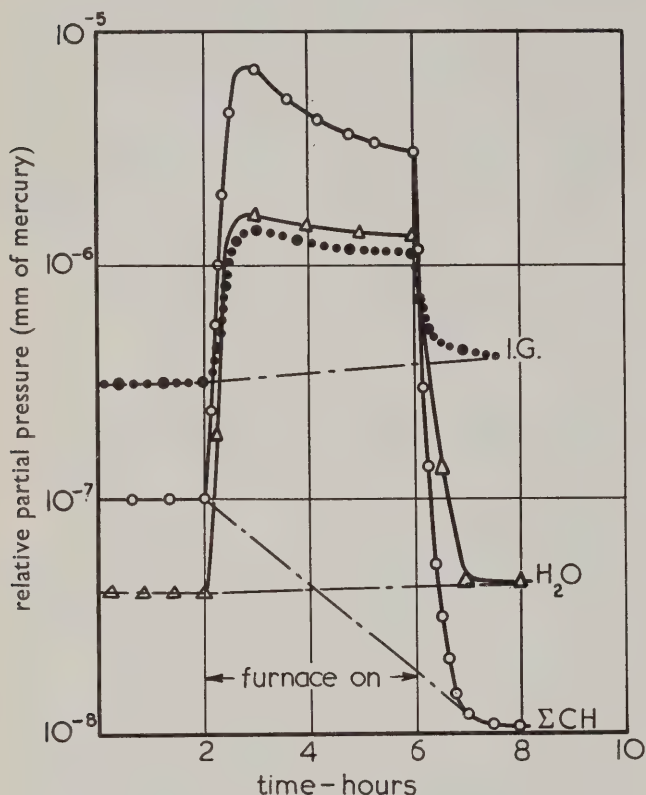
Fig. 3. Changes of partial pressures during baking at 100° C.

period of 4 h. The cold trap temperature was maintained at − 78° C throughout. The results show that when the tube temperature was restored to its normal value the water vapour concentration was unchanged, the sum of the hydrocarbons was reduced tenfold and the ionization gauge reading had *increased*. One conclusion to be drawn from this experiment is, that water cannot be removed by baking if a − 78° C cold trap is present; the water is merely transferred to the cold surfaces from whence it re-evaporates. This is a typical case of the "virtual leak,"[7] but from an adsorbed rather than a liquid surface.[7] A second conclusion is that removal of the hydrocarbon is relatively easy because baking forces the hydrocarbon vapours from the walls to the trap, and a temperature of − 78° C is sufficient to retain them.

(3) ULTIMATE VACUA

It is important to analyse the residual gases at the ultimate pressure after different methods of evacuation because by doing so the origin of the residuals may be established and thus means for producing still lower pressures may be found. Several factors are of importance: (a) the kind of diffusion pump fluid; (b) the temperature of the cold traps; (c) the materials of construction; and (d) the baking temperature. Results for three kinds of fluid, three trap temperatures, two methods of construction and two baking temperatures are given in Table 2.

Table 2 shows that considerable differences in constructional methods and pump fluids had very little influence on the ultimate analyses except where high temperature baking was employed. An unmistakable
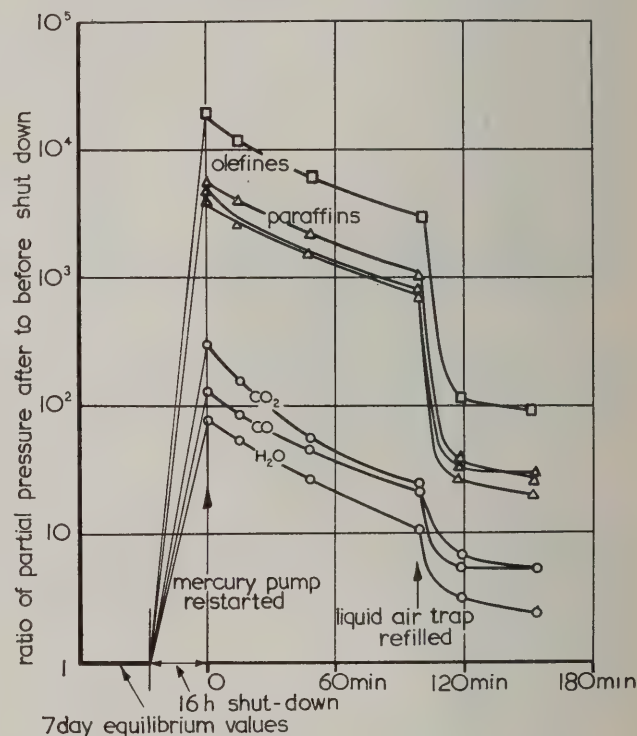
Fig. 4. Diffusion of vapours from rotary pump to high vacuum side. After running at ultimate pressure for 7 days the liquid air trap was removed and the diffusion pump was shut down for 16 h; the rotary pump was left running.

conclusion, therefore, is that baking at the highest possible temperature should be practised. In all trapped systems the most abundant residual (apart from mercury) was water. Residual hydrocarbon pressure was generally higher for oil pumped systems than for those using mercury pumps, but this difference was not marked. (Compare columns (1), (4), (6) of Table 2.)

The presence of hydrocarbon molecules in baked and trapped vessels evacuated by mercury diffusion pumps may be explained in the following way:—

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In the initial stages of evacuation, when the diffusion pump cannot be switched on for fear of oxidation, and the cold trap cannot be cooled because this would condense large quantities of water, oil molecules from the rotary pump diffuse throughout the system. Some of these molecules are adsorbed on the fine side. The difficulty of preventing adsorption (and subsequent re-evaporation) is so great that baking at temperatures up to 250° C for several days, and maintaining the cold trap continuously at liquid nitrogen temperature, seldom succeeds in completely removing the hydrocarbon. Fig. 4, showing the effect of shutting off the diffusion pump and removing the cold trap from a previously

(a)

Fig. 5. Formation of carbon monoxide (a) on a tungsten filament, Curve I—ionization gauge on, Curve II—ionization gauge off. (b) Effect of ionization gauge filament temperature. Partial pressure of water $= 10^{-5}$ mm of mercury

(b)

well evacuated tube, supports this explanation. Similar reasoning will also account for the presence of mercury. For example, as Table 2, column 2 shows, a system baked for many days at 250° C with its mercury pump trapped by solid carbon dioxide still contained mercury vapour at a partial pressure about 10^3 times higher than the vapour pressure of mercury at the trap temperature.

(With liquid nitrogen the disparity is greater, a figure of 10^{10}–10^{14} times being usual.)

The mass spectrometric observations thus support Becker's suggestion that when very low pressures are desired an active carbon trap should be placed between the rotary and diffusion pumps.[8]

(4) FORMATION OF CARBON MONOXIDE

(a) *Steady state.*—To establish the origin of the carbon monoxide observed during the initial evacuation, the following experiments were carried out: (1) the partial pressure of water was gradually increased and corresponding values of the water and carbon monoxide intensities were measured (a) with the ionization gauge working; (b) with the gauge switched off. These results are plotted in Fig. 5(a). (2) With fixed water content the filament current of the ionization gauge was gradually increased, and the magnitude of the carbon monoxide content was plotted as a function of filament current with the result shown in Fig. 5(b).

These two curves together lead to the conclusion that carbon monoxide may be formed on any hot tungsten filament operating in a vacuum system. It is known that tungsten filaments which have operated in hydrocarbon vapour may be converted to tungsten carbide.[9] Thus, the elements required to form carbon monoxide are present in the system. The reaction mechanism, however, appears to be very complex and a detailed explanation will not be attempted in this paper. The important practical conclusion is that if either water or hot filaments

Fig. 6. Formation of carbon monoxide on a "flash filament." Total number of molecules formed per flash after switching off filament for different time intervals. (Water partial pressure 10^{-6} mm of mercury.) The number of water molecule collisions with the filament during the adsorption period is shown in the upper row of figures along the x-axis. Curve I—filament current 1·2 A; Curve II—0·89 A

can be eliminated from pumped vacuum systems, the carbon monoxide content is likely to be very small, i.e. dissolved carbon-monoxide does not readily diffuse out of the metals—brass, copper, stainless steel—or the glassware used in continuously evacuated apparatus.

(*b*) *Formation of carbon monoxide on a "flash filament."*—If a tungsten filament, normally running at a high temperature, is switched off for a short period it will begin to adsorb residual gases. When the filament is switched on again these gases will be desorbed, thus producing a transient increase in pressure. Such a transient has been used in the so-called "flash filament" technique for estimating very low pressures.[10] In view of the evidence of paragraph 4(*a*) it seemed possible that some of the desorbate might be carbon monoxide. Analysis of the gas evolved in flashing a tungsten filament attached to a side tube of the mass spectrometer was therefore attempted. The filament was switched *off* for time intervals of 1, 2, 5, 10 . . . 100 sec and the amount of carbon monoxide generated in the "flash" was calculated from the time integral of the observed increase in the mass 28 peak and the known speed of the pumping system. The duration of the flash was approximately 1 sec.

Results for a typical experiment are shown in Fig. 6, which indicates that for initially clean tungsten one carbon monoxide molecule is formed for every 10 or 20 water molecules impinging on the filament.

A more refined technique, for example, one in which the auxiliary filament was placed inside the mass spectrometer ionization chamber would enable this problem to be studied more thoroughly. The method could be extended to the study of low-pressure adsorption problems in general and would be most helpful where the exact nature of the desorbate or effect producing an observed pressure change is in doubt.[11]

(5) THERMAL DECOMPOSITION OF DIFFUSION PUMP OILS

A lower limit to the safe heat input to diffusion pumps is set by the fact that gases can "back diffuse" through the jet. This effect is selective and is most pronounced for the lighter gases.[12, 13] An upper limit to the heater power is set by the danger of thermally decomposing the oil. Fig. 7 shows the variation with heater power of the partial pressures of hydrogen, water, carbon monoxide and dioxide, gaseous hydrocarbons and air for a metal mass spectrometer pumped with Octoil S. A solid carbon dioxide trap condensed the higher hydrocarbons.

It will be observed that the pressures of hydrogen and carbon monoxide were approximately equal and that each was proportional to the water vapour pressure. It is therefore reasonable to suppose that these gases were formed on the instrument filaments by the process described in paragraph 4. Thus there was no evidence to suggest the thermal decomposition of the oil to hydrogen. On the other hand, the increase in the con-

centration of C_2–C_8 hydrocarbons with heater power was established by the curve marked ΣCH. The inference of this experiment is that the increase in ultimate pressure of oil diffusion pumps which is often observed when they are operated at excessive power input is associated with the formation of lower hydrocarbons rather than of hydrogen, carbon monoxide or dioxide. The very large increase in air pressure at lower wattages was, of course, due to the fact that the backing pressure was not zero.

From the curves in Fig. 7 the factors contributing to the ionization gauge reading, and the usefulness of this gauge in fixing the optimum heat input are quite clear.
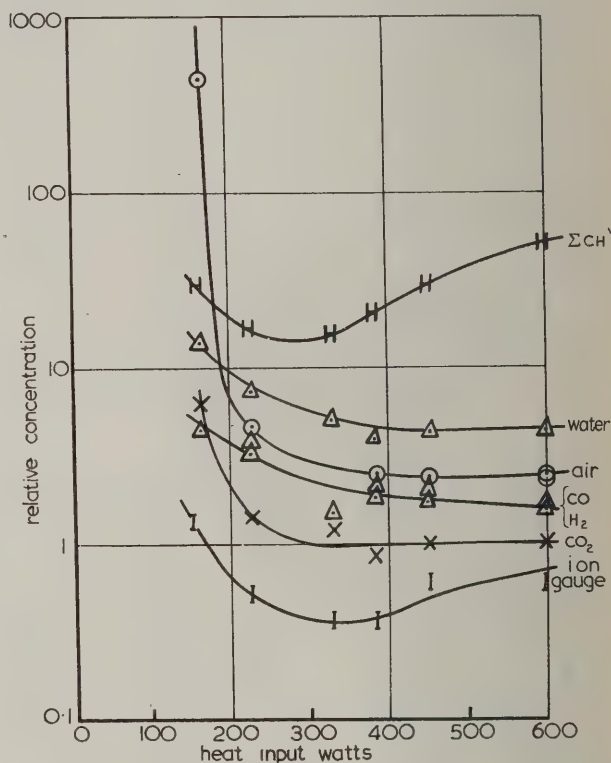
Fig. 7. Thermal decomposition of diffusion pump oil. The results show that the products of thermal decomposition were C_2–C_8 hydrocarbons and that neither hydrogen nor oxides of carbon increased with heat input.

(6) REACTION AND EXCHANGE OF GASES AT SURFACES

It is known that the admission of a gas to a vacuum system will bring about the removal of some adsorbed molecules of other gases and that some molecules of the new gas will be adsorbed.[14] The mass spectrometer enables this process to be studied quantitatively, both with regard to the nature of the surfaces and the gases. The time rate of the exchange during a surface reaction has the general form shown in Fig. 8. This figure shows

a specific case of admission of a halide XF to a system which has not been sufficiently well baked to remove all the adsorbed water. The gross effect is an increase in the total pressure indicated by the ionization gauge. The mass spectrometer shows this pressure rise to be due to two gases; first an initial and rapid increase in hydrogen fluoride caused by reaction of XF with adsorbed water, second a complete absence of XF until most of the water has reacted, then a slow increase in XF.
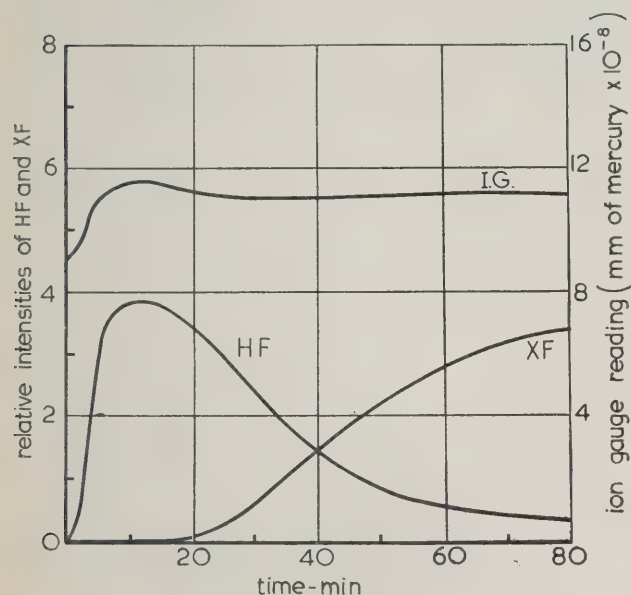
Fig. 8. Reaction and exchange of metallic fluorides with absorbed water. The fluoride reacts with absorbates and the substances initially observed in the gas phase are HF and organic fluorides. The metallic halide itself appears only after most of the absorbate has reacted

Less violent effects can be observed in passage of gases through grease taps. Mettrick and Nicholson[15] showed that if butane was admitted to a vacuum system through greased taps and then pumped away until its concentration was below 0·1% of the maximum value, subsequent admission of argon would re-introduce about 2% of the maximum value of butane previously present. The addition of hydrogen would bring off further butane together with some argon. On the other hand, if gases were handled entirely by mercury cut-offs no evidence of exchange of adsorbates could be found for any of the so-called permanent gases. Strongly adsorbable materials such as alcohols and halogen compounds would exchange to a greater or lesser extent no matter what surfaces were used.

CONCLUSIONS

This paper gives results showing how mass spectrometry may be used for quantitative studies of the nature and behaviour of gases in high vacuum systems.

The experiments described were designed to give semi-

quantitative analytical data on operations of everyday occurrence in laboratory vacuum systems. The examples given are not exhaustive. Many other processes remain to be analysed; for example, the flow of mixed gases; the degassing of gasket materials; the gases evolved in vacuum smelting, sputtering and evaporation; the efficiency of getters. Moreover, all these processes may be studied in greater detail than has been attempted in this paper. Mass spectrometry can, in fact, be applied with advantage to any vacuum problems where the nature of the gas is in doubt, and to others where it is difficult to distinguish between real gas effects and the drifts in gauge readings produced by changes of temperature, work function, accommodation coefficient, or any other physical property upon which the action of the gauge is based.

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DISCUSSION

Mr. F. H. Reynolds: What are the relative merits of the magnetic analyser spectrometer and the radio-frequency mass-spectrometer recently described by W. H. Bennett of the National Bureau of Standards? In view of the bulk of the magnetic analyser, might not an r.f. spectrometer, even if of lower resolution, be more suitable for leak detection?

Author's reply: Figures published by Bennett indicate that by using 1 000 times as many electrons as are used in the older types of instrument, a sensitivity about 2 000 times greater can be achieved. This does not necessarily mean that Bennett's instrument would be sensitive as a leak detector, because in leak detection the central problem is the detection of traces of probe gas in the presence of a high background pressure of air. It is quite possible (*a*) because ions are formed in different parts of a space which is not field free, and (*b*) because of energy losses due to collisions and mutual repulsions in the intense beam passing through the tube, that *some* of the background air ions will arrive at the collector at the same time as the probe gas ions. For the sensitivity to equal that of a conventional mass spectrometer leak detector, not more than 1 in 10^5 of the air ions should do this. So far as I know Bennett has published no figures from which a quantitative estimate of the performance of his spectrometer as a leak detector could be estimated.

If small bulk is desired the tube in a conventional instrument can be made very small. For example, in one commercial leak detector, the tube is about the same size as an ordinary receiving valve, and the magnet weighs only a few ounces.

Mr. W. H. Walton: Is there any difficulty in distinguishing carbon monoxide from nitrogen in the mass spectrometer?

Author's reply: In the work described in the paper there was no difficulty in distinguishing between carbon monoxide and nitrogen because the experimental conditions were such that no significant quantities of nitrogen either from leakage or from surface adsorbates could be present. In ordinary analytical work binary mixtures of these gases can be analysed because the molecules dissociate on ionization, giving ions in the following relative abundances:—

m/e	30	29	28	16	15	14	13	12
CO	0·24	1·27	100	1·84	—	0·83	0·05	5·18
N_2	—	0·74	100	—	0·02	4·32	—	—

Since only the weaker lines are unique, mixtures of these gases cannot be analysed so accurately as gases where the parent molecules have different mass numbers. This is particularly true when only traces of one gas are present in large quantities of the other.

Large-scale vacuum dehydration

By D. R. Goddard, M.Sc., A.Inst.P., British American Research Ltd.,
Wishaw, Lanarkshire

The use of pressures in the micron region in vacuum dehydration has recently extended the field of application of this process. The freeze-drying process for the large-scale preparation of biological substances and the drying from the liquid state of less heat-sensitive materials are illustrations of industrial importance. Freeze drying is not discussed in detail, but recent applications of the liquid film method are shown in a short description of a plant for the dehydration of orange juice and a continuous belt drier for the dehydration of coffee.

The advantages to be gained by the use of low pressure-low temperature dehydration are that low moisture contents may be obtained without oxidation or thermal decomposition with the resultant preservation of the heat-sensitive properties of the original substance.

Evaporation under reduced pressures is a standard practice in the chemical and allied industries as a means of drying, but it is only during recent years that developments arising mainly as a result of the accelerated research demanded by the war, have made possible the use of extremely low pressures for dehydration on a large scale.[1,2]

VACUUM DEHYDRATION

High vacuum dehydration is a technique for the removal of liquids in the vapour phase from products maintained at relatively low temperatures and free air pressures. The technique utilizes the simple fact that liquids boil at lower temperatures under reduced pressure. In the case of water, for example, the boiling point is reduced to $0°$ C at a pressure of $4 \cdot 6$ mm and at $0 \cdot 034$ mm the boiling point is $-50°$ C. Thus water will sublime from the solid state at sufficiently low pressures, a fact which forms the basis of the freeze-drying process so well known in the biological field for the drying of blood plasma, penicillin, and other important biological substances. Drying from the liquid state is undertaken on a large scale at moderate pressures in the millimetre range and will be illustrated by some recent examples in the food industry.

The main advantages to be gained by the use of low pressure–low temperature dehydration are as follows: Materials may be dried to a very low moisture content without oxidation or thermal decomposition. The products may be obtained in a highly porous and dispersable form. It is therefore of importance in the drying of heat sensitive or oxidizable materials and where it is necessary to preserve such materials in a form which is easily reconstituted and which retains as much as possible of the properties of the original substance. Secondary considerations such as the reduction in bulk for transportation and storage purposes are also of importance, especially in the case of food-stuffs from a commercial point of view.

The physical principles involved in high vacuum dehydration are essentially the same as those involved in molecular distillation. Whereas, however, distillation normally involves the separation of liquids of nearly similar boiling points, dehydration is simply the removal of water from a mixture of solids. Again, molecular distillation is applicable more especially to substances of high molecular weight. However, the criteria to be observed in distillation apply in vacuum dehydration, namely, the need for low impedance paths between the evaporating surface and the condenser, good heat transfer conditions and low air pressures to allow free evaporation.

In drying any products, in particular diluted solutions, it is usual to preconcentrate the solutions by means of vacuum evaporation. In the case of very heat sensitive materials such as drugs, this may not be practicable as they are dried from the frozen state. With liquids having high solids content, however, dehydration may often be carried out at temperatures above the true freezing point of the mixture or solvent. Drying rates can be much greater than with freeze drying and the process can be made continuous by the use of drying from thin films instead of the freeze drying batch method. Whatever the drying process used, the conditions of temperature and pressure must be carefully controlled in relation to the time cycle to ensure the correct product properties.

VACUUM DEHYDRATION SYSTEMS

The essential requirements for vacuum dehydration are a vacuum evaporator and a pumping and vapour absorption system. With the exception of steam ejector systems, the pumping and absorption systems must be separate. The different types of vapour pumping systems[3] are:

(i) Those using chemical desiccants and cold traps (Fig. 1 (*a, b*)) which are essentially static or batch methods, although with mechanically refrigerated cold traps it is possible to collect up to 50–100 lb of ice at a time and continuous drying cycles are made possible by the use of multiple traps with provision for melting the ice formed at intervals during the cycle.

and such systems have been used for drying penicillin and other heat sensitive materials on a large scale.

(iii) The continuously operating cold trap (Fig. 1(*d*)) is generally used at pressures below 500 μ and has proved successful in large-scale freeze drying of penicillin, streptomycin, and the freeze drying of blood plasma. In principle, the vapour is condensed on the inside wall of a refrigerated cylinder and is scraped off by a rotating scraper blade as flakes of ice. The ice is collected and removed at intervals without interrupting the drying cycle. The air is removed by a diffusion pump. It is possible to condense up to 55 lb of water vapour per hour with a large unit. Generally, however, a batch of smaller units are used in parallel.

(iv) A continuous version of the chemical desiccant method is the liquid absorption system[4] shown in Fig. 1(*e*). The air is removed by a rotary vacuum pump to a pressure between $\frac{1}{2}$ to 4 mm (these pressure limits are mainly for economic reasons). The water vapour is absorbed in a solution of cooled lithium chloride or other hygroscopic medium. The absorbed water is boiled off and the re-cooled lithium chloride is circulated again into the absorption column. Plants have been designed to deal with up to 200 lb of water per hour.

The design of the drying chambers or evaporators

Fig. 1. Schematic diagrams of five representative types of high vacuum drying systems

(ii) The steam ejector shown in Fig. 1(*c*) is the only method in which the air and water vapour are removed by the same process. If adequate steam and cooling water are available, it forms the most economical method of drying at moderately low pressures, say above 1 mm total pressure. With the use of a four- or five-stage ejector, air pressures as low as 25 μ can be attained

Fig. 2. Process flow diagram of orange-juice dehydration plant of Vacuum Foods Corporation (Florida)

calls for special consideration in relation to the material to be dried. Heat transfer and diffusion conditions, impedances and drying rates are of a somewhat different nature to those met with at higher pressures. If batch
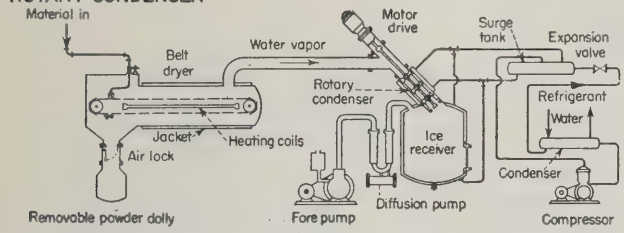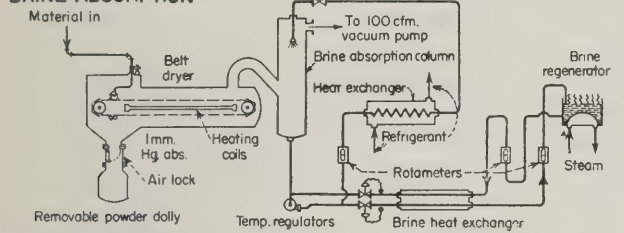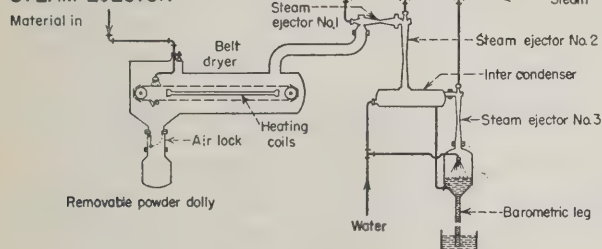
Reproduced from Food Industries

Fig. 3. Belt-drying vacuum systems

Foods Corporation used in America in the orange-growing areas of Florida for the drying of orange juice.[5] The complete plant is illustrated in the flow diagram, Fig. 2. In its original form the plant provided for the concentration of orange juice in falling film columns and then complete drying to a powder at a lower pressure in other columns with refrigerated walls on the rotary condenser principle. For economic reasons the powder is not now produced and the concentrated form (50% solids) is frozen and marketed through the frozen food distribution system on a very large scale. This single plant processes 75 000 gal of juice per day and indicates the growing importance of this type of vacuum technology.

THE BELT DRYER

An alternative method for the drying of liquids from thin films under high vacuum, is the so-called belt dryer, developed by the National Research Corporation in the U.S.A.[6] In principle it consists of a continuous horizontal conveyor belt in a vacuum chamber on to which the material to be dried is sprayed. The water is boiled off with the help of heat from steam coils between the belts and is removed by one of the three methods illustrated in Fig. 3, depending on the pressure used. The solution is preconcentrated before being sprayed on to the belt. On its passage round the belt the solution rapidly dries in the form of a froth which clings to the underside of the belt, where it is scraped off and collected through an air lock in a removable container. The product is obtained in a pseudo-crystalline flake form which has highly lyophilic characteristics.

A comparison of the different drying systems is given in the table:

Table 1. *Comparison of drying systems*

Based on a belt of 40 ft^2 area and a typical solution with 35% solids

	Belt dryer with rotary condenser	Belt dryer with brine absorption system	Belt dryer with steam ejector system
Belt temperature (° F)	0 to 100	40 to 160	80 to 200
Operating pressure (mm)	1×10^{-3} to 0·5	0·5 to 4	> 4
Dryer capacity (dry material lb/h)	5	10	20
Water condensed (water lb/hr)	9·3	18·6	37

methods are used, either for freeze drying or liquid drying, thin layers of material allow rapid heat transfer and diffusion of vapour through the product. The difficulties of obtaining good heat transfer and diffusion of vapour are reduced by the introduction of the liquid products in the form of thin films on vertical wall dryers or on to continuously moving belts. The foremost example of falling film drying under high vacuum is the recent application to the concentration and drying of orange juice.

DEHYDRATION OF ORANGE JUICE

Many attempts have been made to produce a satisfactory concentrated or dried form of fruit juices. One of the most successful has been the process of the Vacuum

The largest plant so far envisaged has a belt area of 400 ft^2 and will remove 200 lb water/hr. The first application of this type of plant was for the drying of coffee extract to produce a form which was readily soluble. Milk and amino acids are among other substances which may be dried in this way.

The belt dryer is especially suitable for drying at very low pressures and may find application for continuous freeze drying.

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DISCUSSION

Mr. I. Varnam: Are there any economic and performance criteria from which the choice of type of pump (steam ejector or mechanical) and trap system can be made? Also, do temperature and degree of vacuum affect only the rate of drying, or do they also affect the ultimate equilibrium? Has Mr. Goddard any experience of the "air-flow" and "cyclic" processes described in Bell Monograph B.1517?

Author's reply: A series of curves presented by Schwarz and Penn[1] and reproduced here show the relative cost of water removal at different pressures for various pumping systems. At higher pressures mechanical pumps are undoubtedly the most economical for the removal of moderate quantities of water (a few pounds per hour). For large systems, however, the steam ejector is more economical, but a limitation is that it becomes difficult to run at pressures much below 1 mm. Below this pressure the vacuum diffusion process using cold traps, either static or continuous, depending on quantities handled, becomes the only practical one apart from chemical absorption methods, which are limited in capacity. The cost of drying increases approximately exponentially with decreasing pressure, and so it is desirable to dry at the highest pressure and temperature consistent with the retention of the quality required in the finished product.

According to Flosdorf,[2] this final temperature to which the product may be taken, and the vapour pressure of the product at that temperature, have a bearing on the temperature required for the condenser or the pressures required in direct pumping. The latter determine the final moisture content or the ultimate equilibrium. Whatever the vapour pressure of the dry product the condenser or pump must establish a pressure which is lower. With a cold trap at $-40°$ C a vapour pressure of $100\,\mu$ is easily obtained, and it is rarely that products are encountered with vapour pressures less than $200\,\mu$ at $50°$ C when containing 0.5% moisture. With higher temperatures lower moisture contents may be obtained.

I have no direct experience of the "air-flow" and "cyclic" processes. In vacuum dehydration it is essentially a utilization of the vapour pressure of the water itself that constitutes the driving force, rather than the use of air as a carrier either by means of forced draft as in the "air-flow" method or by means of an exchange principle as in the "cyclic" process. Textile drying, which is, I understand, the subject in which Mr. Varnam is particularly interested, can be carried out at much higher temperatures than those needed for heat-sensitive materials, such as penicillin or fruit juice, and at pressures
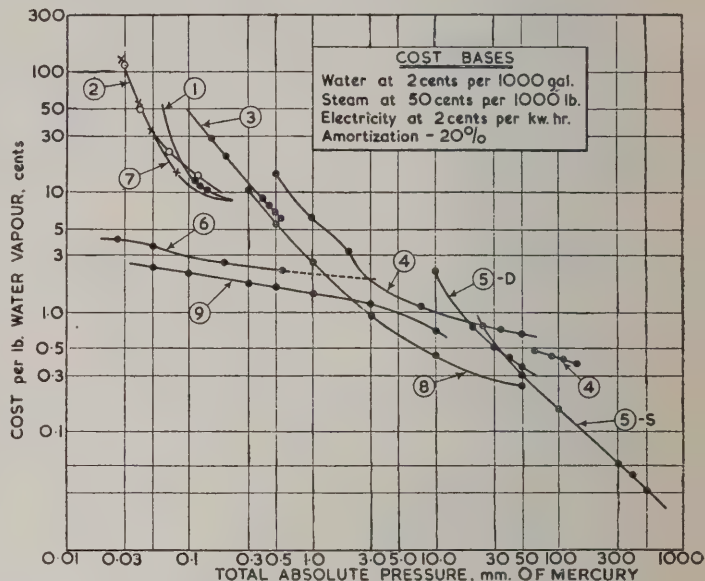
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Representative cost data for various water vapour pumping systems. Investment charges include pumping units as installed, exclusive of driers or utilities

(1) Steam ejector, 3 (18 in., 4-stage) in parallel. (2) Ditto, 10 in, 5-stage. (3) Ditto, 14 in., 5-stage. (4) Ditto, 2- and 3-stage. (5) Single- and double-stage mech. vacuum pumps. (6) Vacuum diffusion process. (7) Steam ejector, 2 (24 in, 5-stage) in parallel. (8) Ditto, 3-, 4-, and 5-stage. (9) Chemical absorption systems.

of the order of millimetres rather than microns. Further, the use of high vacuum for textiles is probably ruled out by the relatively high cost per pound of water removed compared with the unit cost of the material. A very efficient method for textiles, described by Rabold,[3] uses a drier fired by vaporized oil burners, air for combustion being supplied by fans. The products of combustion mixed with superheated steam are recirculated between drier and combustion chamber 24 times per minute.

From the point of view of vacuum dehydration in general the air-flow and cyclic drying processes are of great interest as possible auxiliaries to the high vacuum drying process for the purpose of pre-concentration or partial drying, which can usually be carried out at higher temperatures and pressures.

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The vacuum system of the Birmingham proton synchrotron

By L. Riddiford, M.Sc., Ph.D., A.Inst.P., Physics Department,
The University, Birmingham

The theoretical requirements of the vacuum system of the Birmingham $1 \cdot 3 \times 10^9$ eV proton synchrotron are considered, and the design and development work performed to satisfy these requirements described in some detail. To be certain that no difficulty in operating the accelerator will be encountered because of gas scattering of the proton beam by the remaining molecules, it is necessary to produce a vacuum of about 1×10^{-6} mm of mercury air equivalent in the extended vacuum chamber. The vacuum chamber has to be in the form of a hollow ring about 30 ft in diameter, with a rectangular cross-section 40×10 cm. Because of the high degree of vacuum required, and the fact that a heavy hydrocarbon molecule "gas-scatters" very much more than does an air molecule, the problem of the ultimate vacua of oil diffusion pumps is of the greatest importance. An attempt is made to throw some light on this difficult subject.

There is not space to trace the historical development of particle accelerators. However, it can be said with safety that the work in this field over the last twenty years would have been impossible without the suitable application of modern vacuum techniques. The splendid technical achievements of Lawrence[1] with his cyclotron, and Kerst[2] with his betatron, which have led firstly to the many giant synchro-cyclotrons in operation or under construction to-day, and secondly to the proton synchrotron of Oliphant,[3] have relied in the first place upon the provision in all cases of a suitable vacuum space in which the particles are accelerated.

The heart of all accelerators is the vacuum box, and only the use of the latest techniques has permitted experimenters to evacuate these vessels to the pressure required, ranging from 10^{-4} to 10^{-6} mm of mercury. Development in vacuum engineering has in large part gone hand in hand with accelerator development, for only of late has the chemical industry begun to be interested in large-scale vacuum plants. And, of course, the demands of the research laboratory are most critical; in fact, the stage has now been reached when the requirements of the proton synchrotron are straining present equipment and techniques to their limit. Relatively little experience with vacuum systems of moderate size is sufficient to impress on one the difficulty of achieving, and maintaining, pressures of about 1 or 2×10^{-6} mm of mercury air equivalent, as compared to the relative ease with which it is possible to obtain 1 or 2×10^{-5} mm; although theoretically, in a well-designed system, there appears to be no reason why, with the aid of liquid-air traps, vacua corresponding to about 1×10^{-7} mm should not be obtained, for the air-speed of a pump is constant at all pressures below about 10^{-4} mm.

With present design parameters, the Birmingham synchrotron requires a vacuum of about 1×10^{-6} mm of mercury air equivalent for certainty in operation.

REQUIREMENTS OF THE BIRMINGHAM SYNCHROTRON VACUUM SYSTEM

Degree of vacuum.—An elementary calculation of the distance travelled by the particles in being accelerated is sufficient to make one wonder how some of the present-day accelerators were ever made to work, although of course their successful operation is now an experimental fact, and recently and subsequently a successful gas scattering theory has been worked out.

In the case of cyclotrons and synchro-cyclotrons the particles are accelerated very quickly by large voltages, and further, whilst the energy is low they move very short distances on small circles in the strong magnetic field, in contrast to betatrons and synchrotrons where the energy gain per turn is low, and where also the path length is always long. But in cyclotrons ions are formed with thermal velocities only, whilst betatrons and synchrotrons use a beam of particles initially injected at as high an energy as possible. That gas-scattering may be most serious in proton synchrotrons is obvious at once, since for various technical reasons the acceleration time is of the order of one second, rather than about $0 \cdot 005$ sec as in other accelerators. Consequently the injection energy should be very high, and fortunately this is possible with machines of large dimensions, if the beam is injected from a source external to the magnet.

Nevertheless, for the reasons stated, a theory of gas-scattering in synchrotrons and betatrons has been worked out by Blachman and Courant,[4, 5] with special reference to the proton synchrotron. At the time, application of the theory to existing electron accelerators was not made, but it can be shown that it does agree well with those experimental results that are available.

The effect of a scatter is to set up free oscillations about the equilibrium orbit, which follow the well-known laws first described by Kerst and Serber.[6] The result is that for injection at energy T_i electron-volts on to an orbit of radius R cm, if energy is added at a rate eV electron volts per turn, at a pressure p mm of mercury air equivalent the maximum vertical mean-square gas scattering amplitude is

$$\overline{B^2_{max}} = 8 \cdot 17 \times 10^6 . R^3 p/(eV)T_i \qquad (1)$$

Almost all the scattering occurs when the particles are slow-moving, and the value of $(\overline{B^2})^{\frac{1}{2}}$ given by equation (1)

occurs when the energy has reached four times its injection value.

Now it had been noticed by Lawson,[7] and Elder and others,[8] that if the useful vertical aperture of a betatron vacuum tube is varied by moving an internal shutter, in the form of a wire, there exists in the centre of the tube a minimum useful aperture (of the order of a centimetre
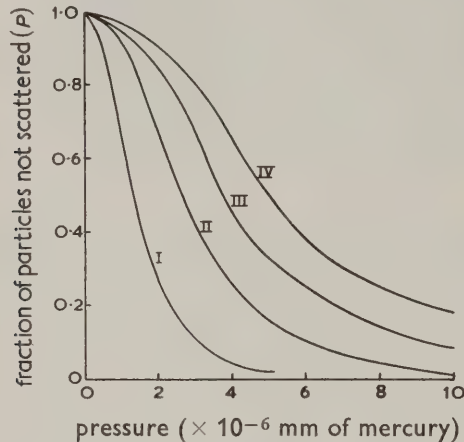
Fig. 1. Fraction P of particles not scattered to the walls of the vacuum chamber, as a function of pressure, for various injection energies, in the case of the Birmingham synchrotron.

Vertical semi-aperture, $A = 5$ cm.; radius of orbit, $R = 450$ cm.; field index, $n = 0.75$; energy added per revolution, $eV = 200$ electron volts. Injection energy (T_i): Curve I, 0.25; II, 0.5; III, 0.75; IV, 1.0 MeV

or so), and for apertures less than this the beam from the machine disappears completely. For greater apertures the output increases very rapidly indeed with aperture. A similar phenomenon has been reported by McMillan,[9] for the Berkeley 330 MeV synchrotron. Also independently various workers[10,11,12] have noticed that the successful operation of a betatron requires vacua of better than about 2×10^{-5} mm of mercury.

If numerical figures are inserted into equation (1), it is possible to form the Table 1, assuming the working pressure in all cases to be 1×10^{-5} mm. It seems fairly definite from this table that there is a positive correlation between the minimum useful aperture and the vertical gas-scattering amplitude.

Table 1. *Comparison of accelerators*

Accelerator	R(cm)	eV(volts)	T_i (volts)	$[(B^2)max]^{\frac{1}{2}}$ (cm)	Minimum useful aperture (cm)
Melbourne 3 MeV Betatron	7·5	20	$1·5 \times 10^3$	1·1	—
Malvern 30 MeV Synchrotron	10	25	$1·0 \times 10^4$	0·6	0·6
G.E.C. 70 MeV Synchrotron	29	200	$3·5 \times 10^4$	0·5	1·0
Berkeley 330 MeV Synchrotron	100	1 500	$8·0 \times 10^4$	0·8	—

Actually the distribution of particles about the root-mean-square value is a Rayleigh one, in the absence of wall collisions, so that it is difficult to see how gas-scattering could cause complete loss of beam, despite the fact that experimental curves of intensity versus aperture are also of Rayleigh form. However, the detailed calculations,[4] taking wall collisions into account, show that there exists a very sharp cut-off at a critical aperture, the physical reason being that a particular particle can wander across the distribution. This, then, would appear to explain the minimal useful aperture,[13] although the fact that the cut-off is entirely complete, according to experiment, must be attributed to insufficiently sensitive detecting equipment, or to some other phenomenon which eliminates the final traces of beam. It is probable that gas-scattering is the main reason for low output from betatrons with small apertures.

Useful data concerning the three proton synchrotrons now under construction[3,14,15] are collected in Table 2.

Table 2. *Comparison of proton synchrotrons*

Laboratory	Final energy $(10^9 eV)$	R(cm)	eV(volts)	Injection energy T_i(MeV)	A(cm)	$[(B^2)max]^{\frac{1}{2}}$	p for $[(B^2)max]^{\frac{1}{2}} = A$
Birmingham	1·3	450	200	0·5	5·0	3·5	$3·5 \times 10^{-6}$
Brookhaven	3·0	1 000	1 040	3·0	8·9	3·0	$3·0 \times 10^{-5}$
Berkeley	6·5	1 525	1 750	10·0	14·6	1·5	$1·5 \times 10^{-4}$
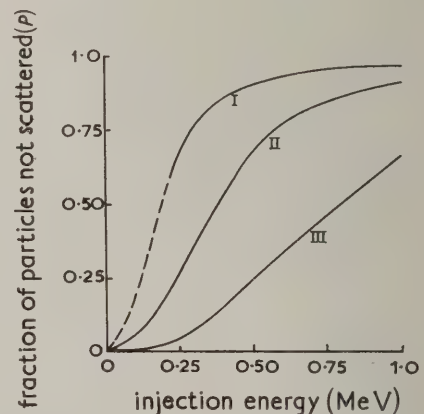
Fig. 2. Effect on the number of particles not scattered to the walls, of varying the injection energy, in the case of the Birmingham synchrotron

$A = 5$ cm; $R = 450$ cm; $n = 0.75$; $eV = 200$. Pressure (p): Curve I, 1; II, 2; III, 4×10^{-6} mm of mercury

Blackman and Courant have shown that, in the absence of other oscillations, and asuming the gas in the vacuum chamber to be air at $300°$ K, the allowable pressure p is

$$p = 0·667 \eta n A^2 T_i(eV)/R^3 \qquad (2)$$

where n is the magnetic field index, A cm is the vertical semi-aperture, and η is a function related to the fraction

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P of particles not scattered to the walls of the vacuum box. T_i and eV are now in units of keV. The results of equation (2) may be plotted in various ways, and this has been done in Figs. 1 and 2, for the Birmingham synchrotron, and Fig. 3 for the Berkeley model synchro-
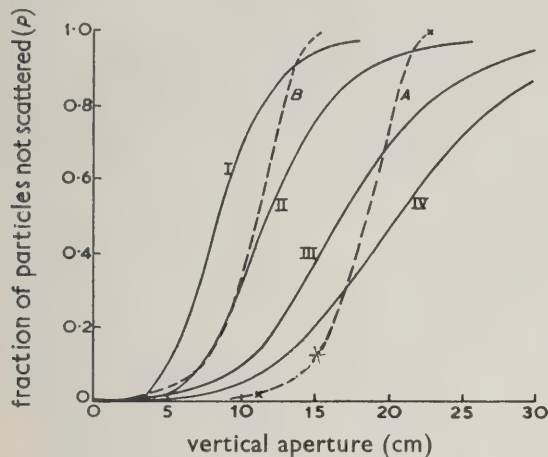
Fig. 3. Curves showing the minimal useful aperture, in the case of the Berkeley model proton synchrotron

Operating pressure $= 2 \times 10^{-6}$ mm of mercury; $T_i = 0 \cdot 6$ MeV; $n = 0 \cdot 75$; $eV = 45$; $R = 350$ cm. Experimental curve A corrected for injection oscillations by displacement to curve B. Pressure (p): Curve I, 1; II, 2; III, 4; IV, 6×10^{-6} mm of mercury

tron.[16] Fig. 1 indicates that part of the curve giving the fraction of particles retained as a function of pressure is of an exponential nature, and there is now experimental evidence for this from two sources, viz. the Malvern 30 MeV electron synchrotron,[17] and the Berkeley 6 MeV model proton synchrotron.[16] Fig. 2 demonstrates the advantage of raising the injection energy (see also Table 2), and Elder and others[18] have presented an experimental curve of similar appearance. Fig. 3 demonstrates the cut-off previously mentioned.

For electron accelerators a detailed comparison of theory with experiment is rendered unsatisfactory because of the presence of unknown injection oscillations and eddy current and other magnetic disturbances. However, if one makes all the correct assumptions it is possible to obtain moderate agreement. In the case of the proton synchrotron the comparison is more clear-cut, and Fig. 3 demonstrates this. Even so an approximate correction for the injection oscillations specified has to be made. The need for restricting these latter oscillations is obvious.

There remains one further matter, arising from the fact that since the slow-moving protons never reach extreme relativistic energies, in the Birmingham synchrotron they are being continually gas-scattered throughout the acceleration cycle. Consequently the beam never becomes small in physical size, and this complicates the problem of extracting it from the machine. Fig. 4 demonstrates the effect. If the whole of the vertical

aperture is used in the initial stages of acceleration, beam size at 1 300 MeV is $2 \cdot 8$ cm vertically and $4 \cdot 8$ cm horizontally, to which must be added a horizontal contribution from phase-oscillations amounting to $0 \cdot 9$ cm.

It is appropriate at this point to mention results obtained by a modification of the above gas-scattering theory to the case of synchro-cyclotrons. The investigation was made following a paper on the 184 in cyclotron,[19] stating that the final beam size is 4 cm vertically and 10 cm radially, of which only $4 \cdot 4$ cm may be attributed to phase oscillations. The reason is not gas-scattering, but neither is it possible to explain the result in terms of magnetic field inhomogeneities or injection oscillations. The answer to this problem is most important from the point of view of Oliphant's cyclo-synchrotron.[20] The calculation does show, however, that at 1×10^{-5} mm the magnitude of the root-mean-square gas-scattering oscillation amplitude can approach
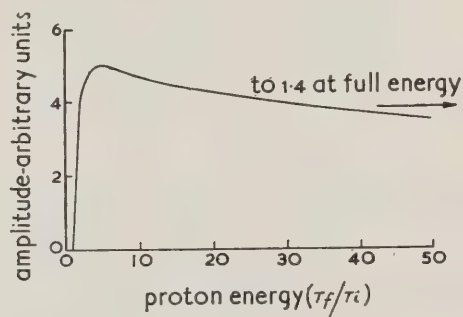
Fig. 4. Instantaneous gas-scattering oscillation amplitude as a function of proton energy, in the case of the Birmingham synchrotron

that of the dee aperture, and so the very best vacua possible should be sought for synchro-cyclotrons also. An increase by a factor of 10 in beam intensity obtained from the 184 in synchro-cyclotron by shaping the central part of the magnetic field,[19] is probably due in part to restriction of the gas-scattering oscillations. In conventional cyclotrons where the dee voltages used are substantially greater, gas-scattering is not important for pressures less than about 10^{-4} mm, because of the more rapid acceleration as the ions are dragged out of the source.

Shape of the vacuum box.—The vacuum box provides a retaining wall which should enclose as much of the magnet air-gap available as possible, since the cost of the magnetic field is the major item in any accelerator which employs one. The enclosure must have sufficient mechanical strength to withstand, with some safety, the atmospheric load, and above all the materials of construction should be vacuum-tight. Eddy-currents, which destroy the shape of the magnetic field, preclude the use of conducting materials in most cases where the magnetic field is a changing one.

The maximum particle energy required, and the cost of the source of power to drive the magnet for providing this energy, determine the size of the box, but apart from this the factors governing shape are the various motions of the particles. For the Birmingham synchrotron a decision was made in terms of a reasonable interpolation from experience with smaller machines, for a gas-scattering theory was not available at that time. The radius of the orbit is 450 cm, and the magnet gap is as drawn in Fig. 5. The useful n-range $(0 < n < 1)$ is a rectangle of dimensions 40×20 cm, but with the present construction only about 40×10 cm is available as usable space. It is of interest and importance to see how this compares with the best theoretical rectangular shape, in view of the substantial difficulties encountered in the manufacture of the vacuum chamber. The manufacture would have been relatively simple with a vessel of simpler shape.
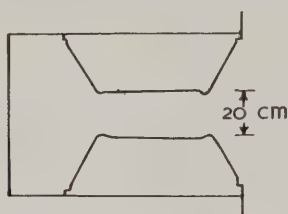
Fig. 5. Birmingham synchrotron magnet air-gap

The deviation of particles from their ideal orbit at the centre of the vacuum box is due to:

(*a*) "Betatron" or injection-free oscillations.

(*b*) Gas-scattering free oscillations.

(*c*) "Spiral" oscillations, arising from the fact that particles of constant velocity, injected tangentially at the outside edge of the vacuum box before the radio-frequency is switched on, will not spiral in smoothly to smaller radii as the magnetic field increases, for their centre of curvature is continually changing.

(*d*) Forced harmonic oscillations, due to inhomogeneities in the magnetic field.

(*e*) Errors in the radio-frequency.

(*f*) Phase oscillations and associated radial oscillations.

(*g*) Other causes such as electrostatic defocusing and space-charge beam-spreading.

It can be shown that in a correct design only (*b*) and (*f*) should matter, although of course the larger the chamber the easier are the many technical difficulties involved. The oscillations from (*a*) and (*b*) are both horizontal and vertical, with amplitudes in the ratio $[n/(1-n)]^{\frac{1}{2}}$, whilst (*e*) and (*f*) cause only radial oscillations. Consequently the ratio of width to height of the box can be written

$$b/a = [n/(1-n)]^{\frac{1}{2}} + \epsilon_1 + \ldots \qquad (3)$$

with $\epsilon_1, \ldots$, the contributions from phase oscillations and other causes. The general synchrotron theory of Bohm and Foldy[21] permits an estimate of ϵ_1 to be made, and in the present case the radial spread from errors in phase is 40 cm. If full use is made of the vertical aperture of 10 cm, then an ideal box would appear to have to be $57 \cdot 3$ cm wide for $n = \frac{3}{4}$. However, since it takes about $0 \cdot 02$ sec for the gas-scattering oscillations to build up to their maximum values, there is this time available for the phase oscillations to be damped. The position is depicted in Fig. 6, indicating that the box needs to be $46 \cdot 6$ cm wide.
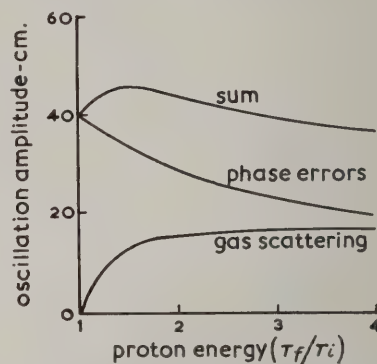
Fig. 6. Instantaneous horizontal oscillation amplitude as a function of proton energy, in the case of the Birmingham synchrotron

A further feature affecting shape is the need to extract the final beam. The energy of the accelerated protons is so great that the beam can only be deflected slightly by whatever method of extraction is employed, and consequently must be brought out at a tangent through a suitable orifice. The only method of doing this is to provide sections of vacuum box which are wider than proposed above, so at two points there are "steps" in the ring which, when provided with suitable orifices, offer means of injection and extraction. Fig. 7(*a*) depicts the end of one of the special sections which have had to be cast to provide such orifices, and also to facilitate reliable vacuum seals to adjacent sections of smaller size.

CONSTRUCTION OF THE BIRMINGHAM SYNCHROTRON VACUUM SYSTEM

The vacuum chamber.—Space is not available to discuss the various possible ways of fabricating the vacuum box. The ring as originally planned was to be constructed in the form of a many-sided polygon from thirty straight sections each of length $12°$ in arc. This corresponds to about one metre. There are pieces of two different sizes to make extraction and easy injection possible, and more than half have four 4 in internal diameter side tubes on them to facilitate speedy pumping and access into the

vacuum chamber. Fig. 7(b) is a drawing of a large section, as originally manufactured in stoneware.

The dimensional requirements of these bodies present a most severe problem indeed to the manufacturers, and the synchrotron project acknowledges the help of Messrs. Hathernware Ltd. of Loughborough who manufactured the vacuum box. Pieces of satisfactory shape have been produced, and the large wall thickness (3 cm) provides a reasonable safety factor in strength of better than 1·5. But chemical stoneware is quite porous, and experiments to correct this porosity were only persisted with in view of the fact that it has only recently been possible to manufacture in an alternative material. It was soon realized that a leak-rate per section of less than 0·2 l/sec at 10^{-3} mm of mercury was desirable with the pumping speed available. Three separate batches of stoneware vessels have been tested, the leak-rate being consistent within groups and averaging out at 20, 1·5 and 180 μl/sec respectively.

Normal leak-hunting techniques are not applicable since it takes some hours for a molecule to pass through the wall. Further, the difficulty of correcting the trouble is apparent when it is considered that the entire leakage into a section may be through a solitary hole of about 0·002 in bore passing straight through the wall. It is likely that the individual pores are not much larger than the size of an air molecule. After much effort it has been found possible to impregnate successfully with a mixture of hot paraffin-wax and beeswax, and the leak-rate in one piece has remained steady at 0·03 μl/sec for many weeks. However, meanwhile Messrs. Hathernware Ltd. have succeeded in slip-casting a body in electrical porcelain supplied by Messrs. Taylor, Tunnicliff and Co., Ltd., and in view of the many disadvantages associated with the stoneware, production of a complete ring in this material is in progress. High-quality porcelain has a porosity at least an order of magnitude below the design figure. However, because of the increased distortion incurred in firing the porcelain, the length of the sections has had to be reduced to one-half that shown in Fig. 7. This means that sixty chambers and sixty seals are now required. The shape of the porcelain half-section is just as excellent as that of the full-length stone-ware pieces. Manufacture in fused quartz and thin 0·007 in stainless steel sheet has been pursued, but the prospect of success in neither case looks hopeful. The Thermal Syndicate Ltd. have succeeded in blowing two silica vessels of moderate shape, but side-tubes would have to be attached in a separate operation, and further the tensile strength of quartz is substantially below that required in practice.* The thirty completed sections have to be smooth-ground very precisely in angle (to ± 0·002 in across an end), so that the ring

fits neatly together. The grinding problem is a most difficult one, but has been solved by the use of suitable jigs.

General layout.—The general plan of the vacuum system is shown in Fig. 8, with an elevation of the arrangement of a pumping manifold near the magnet in Fig. 9. Figs. 10 and 11 are photographs of manifolds and pumps on test, and Figs. 12 and 13 indicate how the sections will appear when in position between the pole-tips of the magnet.

The system was designed fundamentally on the assumption that five or six 16 in diffusion pumps would be employed, backed by a similar number of Kinney VSD 8811 mechanical rotary pumps. There must be some form of manifold connecting the vacuum box to the diffusion pumps, and apart from questions of accessibility, the feature to be kept in mind in designing the manifold is that the vacuum box of the synchrotron
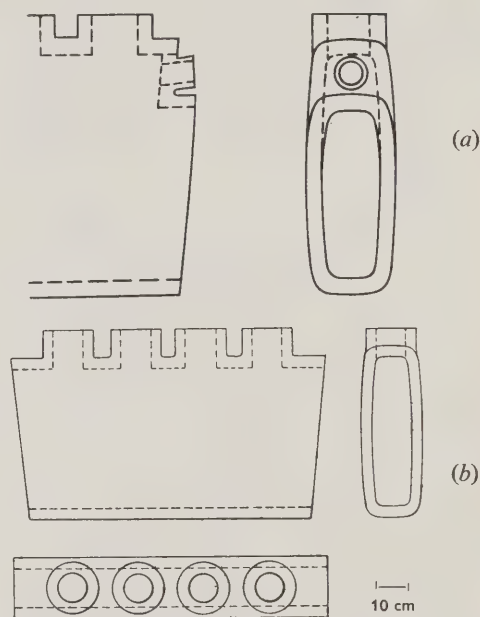
Fig. 7(a). End of special section for injection and extraction of the beam. (b) A section of the Birmingham synchrotron vacuum chamber

itself forms part of the pumping line. For accessibility, and to leave room for beam monitoring, radio-frequency connections, etc., some sections are left free from pumps. That a good deal of care had to be taken in laying out the system is clear from the following figures:

Speed of baffled pump = 2 000 l/sec.
Speed of one section of vacuum box = 1 500 l/sec.
Speed of four ports = 2 000 l/sec.

It is clear, then, that the ring must be pumped at many points to reduce loss of pumping speed and to prevent the occurrence of severe pressure gradients along the vacuum box. The final scheme adopted uses five

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* A rectangular chamber of sufficient strength has now been constructed from slabs of silica 35 mm thick, cemented together with the thermo-setting plastic Araldite. Only one small leak was found in 15 ft of seam. However, for various reasons we are proceeding with the porcelain project.

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manifolds placed one at the top of each of five pumps, each manifold extending over three sections of the chamber, and linking up with twelve ports. The manifolds are made in stainless steel so as not to affect the magnetic field, and were manufactured by the London Aluminium Company, Ltd. They are not placed quite uniformly around the synchrotron, but rather at points where gassing is most likely to occur (at ends of cee and near injector). The ports on the manifolds connect with those on the vacuum chamber through sixty 4 in stainless steel seam-welded bellows. Bellows 3 in long, using flanges with large bolt-holes, provide ample flexi-

Fig. 10. A pumping manifold on test with a 16 in diffusion pump and Kinney mechanical pump, type VSD8811

Fig. 8. Layout of Birmingham synchrotron vacuum system
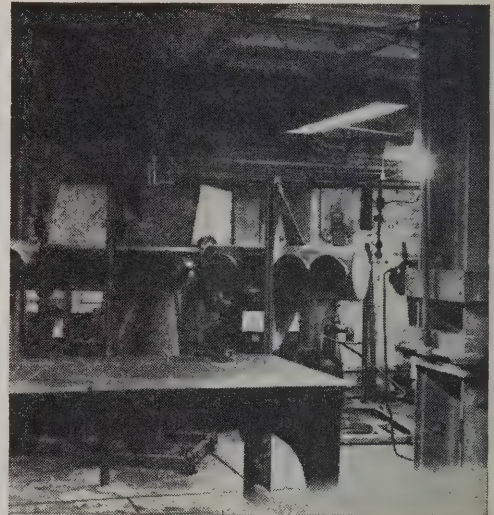
Fig. 11. Front view of pumping manifold with side-arms

bility in all directions for the assembly of such a complicated system, which would otherwise be most difficult, particularly on account of the variations which occur in the manufacture of ceramic materials.

Since the gas-scattering law is an exponential one, local areas of high pressure from leaks are to be avoided. Due to differential pumping, should a single leak occur diametrically opposite a gauge, the pressure reading at that point will be about 400 times greater than the pressure recorded by the gauge. Consequently eight ionization gauges are suitably placed around the ring to present a true picture of what is going on, and incidentally will afford a means for locating the general position of serious leaks.

Fig. 9. Elevation showing magnet, vacuum chamber, stainless-steel pumping manifold with suspended liquid-air trap, 16-inch diffusion pump, and backing line. Flexible stainless steel bellows 3 in long and 4 in internal diameter, are to be connected between ports on the vacuum chambers and ports on the manifolds

The hot tungsten filament ionization gauge is the most simple, robust, and least expensive pressure-recording device available, and despite its known uncertainties is universally employed on any equipment of appreciable size. In small systems where very low pressures have to
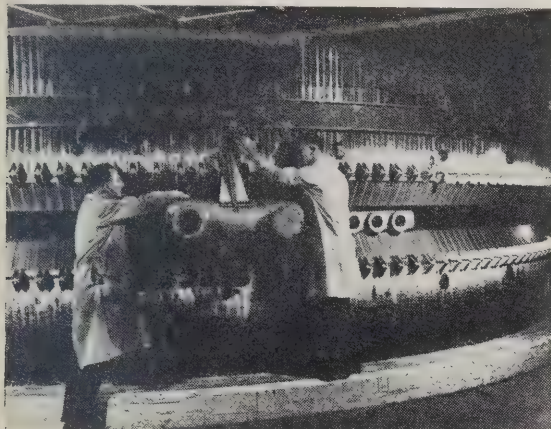
Fig. 12. Birmingham synchrotron magnet with a section of the vacuum chamber and portion of the pumping manifold in place

be known accurately and absolutely, the Knudsen gauge is the only one with sufficient sensitivity. The synchrotron vacuum system demands a combination of these requirements. Since the ion gauge will satisfy all if doubts about its reliability are removed, and in view of the suspicion with which many experimenters regard the gauge, a careful study has been made of its behaviour.
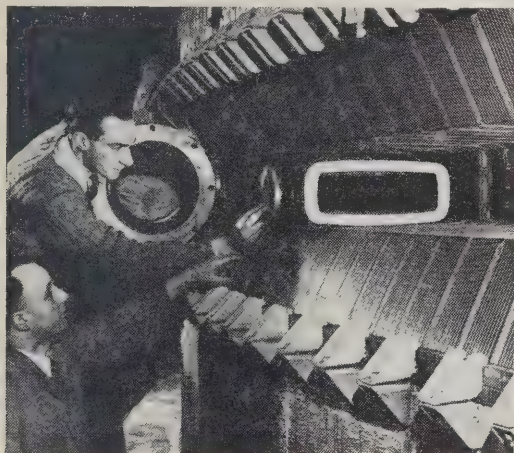
Fig. 13. Close-up of magnet air-gap and section of the vacuum chamber

The result obtained is the satisfaction of its reliability down to the very lowest pressures available.

The fore-vacuum is provided in a 4 in copper line running around the magnet near floor-level. The five rotary pumps are stationed some 30 ft from the magnet,

and link up to the 4 in pipe through a 6 in connecting pipe and metal bellows. A by-pass line connecting up with the back of a manifold facilitates adjustment of the apparatus without switching off diffusion pumps.

Some details of construction.—A feature of all accelerators is the need for a conducting lining inside the vacuum box to remove stray charges from the walls. In the present case two proposals have been investigated. One is that the lining should be in the form of removable mechanical inserts of 0·018 in inconel sheet, as in Fig. 14, saw-cut suitably to reduce eddy-currents. Sample liners have been manufactured to excellent shape. This arrangement has many advantages over the normal technique of chemical coating, but the dangers of field penetration through the saw-cuts, and of short circuits across saw-cuts, leading to eddy-current field troubles, appear to be real ones. The most serious disadvantages with the liners are firstly the difficulty of storing and handling them without damage to the shape, and secondly the fact that the vacuum chambers are not uniform in size. Experiments to see if it is possible to cover all of the extensive inner surface of a section effectively with a chemically applied layer of platinum of resistance about one ohm per square, have been very successful. In view of conflicting reports from different laboratories, the

Fig. 14. Inconel liner to remove charges lost to the walls

importance of bare patches on the surface is uncertain. Therefore to be sure that no such bare patches will be present, the chambers are first glazed with a conducting glaze of resistance one megohm per unit square, and then platinized. It is certain that a combination of conducting glaze and platinum film will provide a perfect screen. Whether platinum or inconel is used, the conducting glaze is necessary. It has the advantage that gaps for the radio-frequency accelerating voltage can be left uncovered without fear of a build-up of charge. Further, certain sections of the film (which act as Faraday cages for monitoring the beam), can then be easily isolated from the rest of the film through the relatively high resistance glaze. The construction of these cages is greatly simplified by the use of platinum rather than inconel. We are indebted to Messrs. Taylor, Tunnicliff and Co., Ltd., for the development of the conducting glaze and platinum technique.

That satisfactory vacuum joints are an essential feature of design is evident when it is pointed out that there are some five hundred of these to be made. The most difficult ones are those between sections of the vacuum box, which must be reliable and easily tested for the reason that the back of the box is inaccessible when assembled. A suitable arrangement consists of four

$\frac{3}{16}$ in $\times \frac{3}{16}$ in soft rubbers, bonded two on each side of a metal plate providing the mechanical strength, as in Fig. 15. It also permits easy electrical connection through the wall of the vacuum chamber. The spring contacts shield the rubber from the beam, and facilitate easy connection to the platinum film inside every chamber. All other gasket seals are double ones also, to facilitate leak hunting and provide means of correcting the effect of a damaged gasket without dismantling the system. The entire ring is forced radially inwards with suitable clamps through a distance of $\frac{3}{4}$ in before evacuation is commenced, so that no subsequent movement occurs as the gaskets compress. It is important to notice that the sections with ports and bellows would, in the absence of clamps, tend to move outwards when the vessel is evacuated, because the force which would normally be on the ports is taken by the manifold, which is rigidly attached to the magnet structure. Cast inconel flanges cemented on to the porcelain ports permit compression of the gaskets between manifolds and vacuum box.

Vacuum valves consist on the backing side of machined 2 in water valves, and on the fine side of 16 in plate valves mounted on the manifolds. Wilson or O-ring seals with double gaskets provide sliding seals. A $1\frac{1}{2}$ in two-way straight-through valve as in Fig. 16 isolates the main vacuum system from the injector vacuum system. The principle of the valve is evident from the schematic figure. A cam with the driving shafts $\frac{1}{8}$ in off centre
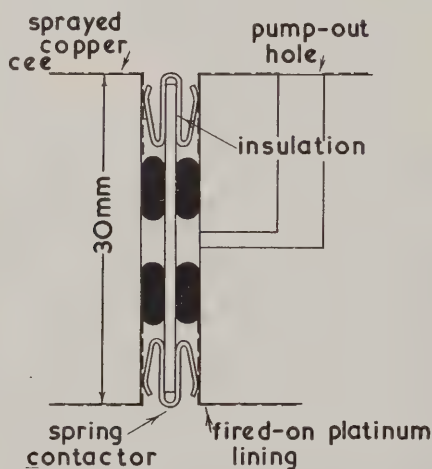
Fig. 15. Gasket for sealing vacuum-chambers together

provides ample sealing pressure, and the device has the advantage of being self-locking. At a number of points around the ring, air-locks are placed to permit introduction and extraction of probes, targets, etc., without disturbing the vacuum.

Leak-hunting is performed using an ionization gauge with a wide-bore connecting tube, and a sensitive galvanometer. The best probing material is liquid trichlorethylene, applied with a painter's brush, the amplification factor over air in this case being of order 25. No stabilization of the circuit is necessary, the method being so extremely sensitive that fluctuations in the ultimate

pressure of the pump are relatively very small. It is possible to detect leaks of order $0 \cdot 01 \ \mu l/sec$ by this method. One manifold (volume 600 l) was assembled, leak-tested, welded, and re-tested to better than $0 \cdot 1 \ \mu l/sec$ in a period of less than four normal working days. It is possible to re-weld all but the largest holes, with the vacuum applied, without danger of the wall in the vicinity collapsing.

A water-cooled staircase-type baffle is inserted into the throat of each pump. Water-cooling is sufficient to keep the oil in the pump, but whether provision of refrigeration-cooling to lower the ultimate vacuum is necessary and
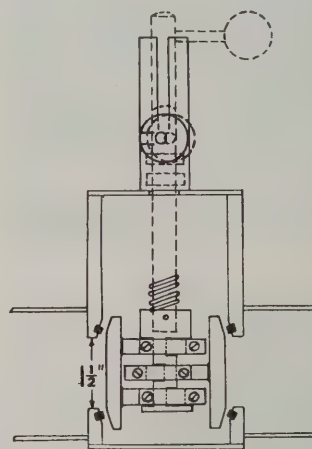
Fig. 16. Injector two-way straight-through isolation valve

worth while is a matter for experiment. Three stainless-steel liquid-air flasks of total volume $7 \cdot 5$ l are suspended in each manifold (Fig. 9) in case they are needed. These increase substantially the pumping speed for the ultimate molecules, and so lower the ultimate pressure. But it is to be noted that the use of liquid-air would cost the Department £20 per 24-hour working day.

Pressure control of the five diffusion pump heaters is obtained with the out-of-balance current from a Pirani-gauge bridge, the gauge being placed in the common backing line. The current operates a Weston relay Model S54, which in turn actuates five Donovan heavy-duty relays type R 920 S, via a suitable Post Office relay. The device results in automatic on-off action as the pressure varies about $100 \ \mu$, with a reproducibility of about $\pm 5\%$.

Water switches consist of simple overflow devices which use the conductivity of the water itself to provide sufficient current to actuate a Post Office relay. Their advantage is the absence of moving parts.

The injector vacuum system cannot be considered separately, for there must of necessity be a connecting hole. The lay-out is as in Fig. 17, with the 500 kV injector tube quite conventional except that the beam moves horizontally instead of vertically. Since the air-pumping speed of the conical tube is relatively low (100 l/sec), because of differential pumping, there is little danger that with reasonable care the main vacuum will be affected by conditions in the injector system.

THE ULTIMATE VACUA OF OIL DIFFUSION PUMPS

Vapour pressures of diffusion pump oils.—Although it is known that the vapour pressures of the better pump oils, at room temperature, are in the region of 10^{-8} mm of mercury, it is not possible to obtain pressures (measured with an ionization gauge calibrated for air) of less than about 1×10^{-6} mm of mercury, and the sensitivity
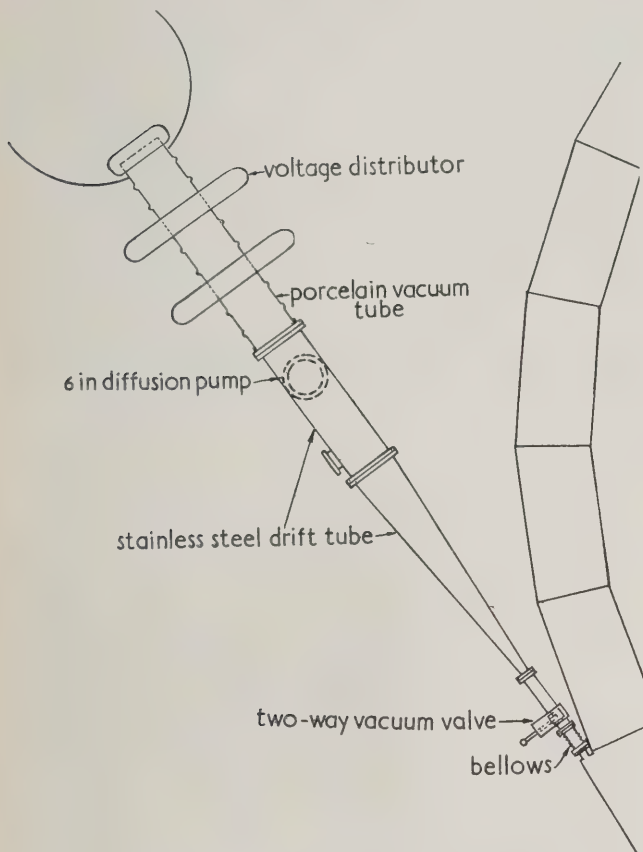
Fig. 17. Injector vacuum system

factor of the gauge for most oils (or rather their decomposition products) is only about 5.[22,23] The conclusion is, then, that the ultimate pressures arise from some other cause, the obvious one to assume being thermal decomposition of the oil.

Nevertheless, the vapour pressure is an important quantity, for a reliable working rule is that oils of low vapour pressure result in good ultimate vacua. There appear to exist only two direct methods of determining the vapour pressure of an oil at room temperature, the first one being the molecular dew method of Kapff and Jacobs.[24] Upper limits to the values of the vapour pressures of the oils octoil S (dioctyl sebacate), Apiezon B, and silicone fluid DC703, have been measured with a similar apparatus to that of Kapff and Jacobs, the results being 2×10^{-9}, 1×10^{-8} and 3×10^{-8} mm of mercury respectively, at 25° C.

The apparatus is drawn in Fig. 18. Of the molecules emitted by a hot oil surface, only those in a preferred direction are allowed to condense on a cold front-silvered mirror, from which, however, they can then evaporate in all directions. If the temperature of the hot surface is slowly raised until a visible film appears on the mirror, it is known that

$$GP_B T_B^{-\frac{1}{2}} \geqslant P_M T_M^{-\frac{1}{2}} \qquad (4)$$

where T is the temperature, P is the vapour pressure, and G a geometrical factor taking account of the collimation of the molecular beam. The ratio P_B/P_M follows at once from equation (4), and P_B is known by direct weighing at elevated temperatures.

The difficulty with the direct weighing experiments, or the alternative boiling point method of Burrows,[25] is that it is not certain that only oil molecules are being boiled off. In both cases extrapolation to room temperature is made by using the Clausius–Clapeyron equation and the usual $lnp - T^{-1}$ diagram. Dushman[26] has presented a summary of the results of this work, and some are plotted in Fig. 19 together with the author's results. It is clear that there is a marked deviation in slope and absolute magnitude from curves obtained with
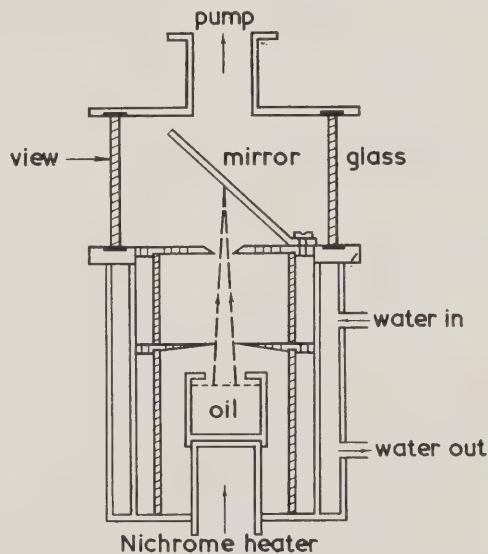
Fig. 18. Molecular-dew apparatus

the dew apparatus. An explanation lies in the fact that hot pump oil is known to decompose readily, and the curves differ in a manner qualitatively in agreement with this assertion. Consequently the value of experiments at elevated temperatures is doubtful, unless conditions compare specifically with those in an operating pump. Similarly, the present dew method is also in error, but not so much, and in this case the slope of the curve at least is probably correct.

A point most important in pump baffle design, which appears in Fig. 19, is that a change of a factor of 2 in vapour pressure arises from a change in temperature of only 3° C.
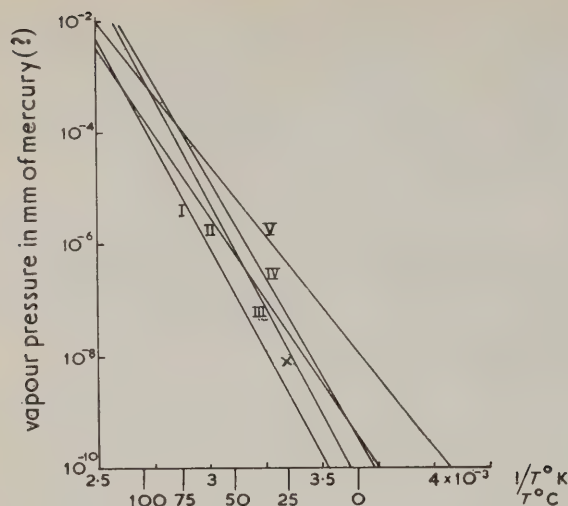
Fig. 19. Vapour-pressure (?) versus temperature diagram for various pump oils

Curve I, Riddiford—Octoil S; II, Dushman—Octoil S; III, Riddiford—Apiezon B; IV, Riddiford—DC 703; V, Dushman—Apiezon B; Point ×, Kapff and Jacobs—Octoil S

mainly of oxygen and carbon atoms, and have molecular weights ($\simeq 500$) about sixteen times that for air, we can say that the scattering effect of 1×10^{-6} mm of mercury of oil vapour is the same as $1 \cdot 6 \times 10^{-5}$ mm of mercury of air.

Consequently for 1×10^{-6} mm of mercury air equivalent, the partial pressure due to oil vapour must be no greater than about 6×10^{-8} mm of mercury. This is in excess of the figures quoted above (except that of Filosofo and others), and the use of traps and baffles will improve the position further. But a more exact knowledge of the nature of the remanent molecules is desirable. One pertinent experimental result from Berkeley[9] is the fact that the effect on the beam of the model synchrotron of allowing the pressure to rise by not keeping liquid-air traps in operation, is the same as leaking-in sufficient air to deflect the ion gauge galvanometer by an equal amount. It is known that liquid-air traps remove the ultimate molecules. Now Found and Dushman[28] have pointed out an empirical law to the effect that the sensitivity of an ion gauge varies in proportion to the molecular weight of the gas in it. This

Table 3. *Final pressures for various oils*

Oil	Vapour pressure (25° C)	15° C	Final pressure (air equivalent) 0° C	− 78° C	− 180° C	S(1/s)‡
dioctyl sebacate	2×10^{-9}	$6 \cdot 2 \times 10^{-7}$	$6 \cdot 2 \times 10^{-7}$	$4 \cdot 2 \times 10^{-7}$	$1 \cdot 3 \times 10^{-8}$	$4 \cdot 2$
dioctyl sebacate		$3 \cdot 1 \times 10^{-6}†$	$2 \cdot 8 \times 10^{-6}$	$6 \cdot 2 \times 10^{-7}$	$2 \cdot 4 \times 10^{-7}$	$3 \cdot 8$
dioctyl sebacate		$2 \cdot 0 \times 10^{-5}†$	—	—	—	$1 \cdot 2$
redistilled dioctyl sebacate		$2 \cdot 5 \times 10^{-6}$	—	—	—	$0 \cdot 0$
Apiezon B	1×10^{-8}	$1 \cdot 0 \times 10^{-6}$	—	—	—	$8 \cdot 9$
Apiezon B		$2 \cdot 5 \times 10^{-6}$	$8 \cdot 8 \times 10^{-7}$	$1 \cdot 2 \times 10^{-7}$	$7 \cdot 5 \times 10^{-8}$	$0 \cdot 0$
Apiezon C		$8 \cdot 7 \times 10^{-7}$	$4 \cdot 3 \times 10^{-7}$	$1 \cdot 4 \times 10^{-7}$	$3 \cdot 6 \times 10^{-8}$	$4 \cdot 9$
silicone DC703	3×10^{-8}	$3 \cdot 2 \times 10^{-6}*$	—	—	—	40
silicone DC703		$8 \cdot 4 \times 10^{-7}*$	$4 \cdot 9 \times 10^{-7}$	$2 \cdot 8 \times 10^{-7}$	$1 \cdot 1 \times 10^{-7}$	$2 \cdot 7$

* Same sample in different pumps at a 9-month interval.
† From same drum of oil at 6-month interval.
‡ S is the air equivalent pumping speed of the ionization gauge for the ultimate molecules.

The second method simply involves the use of a Knudsen gauge with and without the presence of an oil sample. Results recently presented by Filosofo and others,[27] show that the vapour pressure of Apiezon B at room temperature is about 4×10^{-6} mm of mercury. The measurement is in violent discord with those already mentioned, but agrees in order of magnitude with the ion gauge ultimate pressure-reading obtainable with a non-fractionating type of diffusion pump, although when corrections for the sensitivity factor of 5 are made the value is somewhat high.

Ultimate vacua.—The importance of this subject is apparent from what has already been said. Further, if the molecules finally present in a perfectly tight vacuum box are not air, but oil or other heavy molecules, at a substantial pressure, gas-scattering will be very great indeed. By Avogadro's rule, for a given pressure the number of scattering atoms must be greater for gases of greater molecular weight, if the atoms are of about the same mass. Since oil molecules consist by weight

would mean a factor of fifteen over air for oil molecules, whereas the observed value for pumps is five. The experimental facts and Dushman's law can all be retained if it is assumed that the oil decomposition products have an "average" molecular weight of about 160. On this basis an air equivalent ultimate reading of 1×10^{-6} mm of mercury is to be considered satisfactory for the present application.

In another paper Blears has reported the results of an investigation of the nature of the ultimate molecules using a mass spectrometer. The result is a continuous spectrum of masses up to a mass-number of about 300.

The final pressures for various oils have been measured using 16 in fractionating pumps (by British American Research Ltd.), and the results are collected in Table 3.
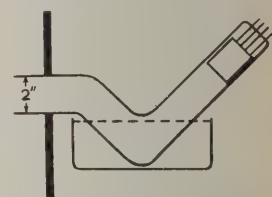
Fig. 20. Arrangement for investigating the effect of refrigeration on ultimate vacuum

All known precautions were taken to obtain what are considered to be true values. The effect of refrigeration was investigated with the experimental arrangement of Fig. 20. The 2 in tubulation overcomes the well-known ionization gauge pumping effects,[29] and a right-angle bend is the simplest type of one-collision baffle allowing maximum tubulation speed. The manner in which the final pressure depends on refrigeration is positive evidence against it being due to oil vapours. In the case of Apiezon and silicone oil at least half, and for octoil almost all the final pressure must be due to decomposition products.

A point of interest arising from the table is that from the point of view of final pressure silicone oil behaves in a similar manner to the carbon oils. This is difficult to understand because of the well-established fact that the "silicone" oils recover rather better than the "carbon" oils from sudden air inbreaks. Further evidence for the fact that it also decomposes is that it produces a vacuum several times better when in a fractionating pump than in a non-fractionating pump. It is now generally accepted that for vacua better than 5×10^{-6} mm fractionating or self-purifying pumps are essential. A further point to be kept in mind is that raising the heater power excessively may seriously worsen the ultimate vacuum. Improvements in performance of oils are possible in two directions: firstly, consistency, and secondly, the production of better ultimate vacua. Table 3 indicates that there may exist serious variations in the properties of oil samples bearing the same name. Further, two samples of dioctyl sebacate extracted from the same drum with an interval of six months between the extractions yielded ultimates of 3×10^{-6} mm and 2×10^{-5} mm respectively, suggesting ageing of the oil. Once a sample has been in use in a pump, deterioration with age may or may not occur, depending on the oil. Whether or not the result for silicone oil, suggesting an improvement with age, is of practical value or not remains to be seen. Perhaps the differences arise from dirt or impurities in the pump, or from minor differences in pump constructional details, which may affect, for instance, heat transfer and consequently the nature of the jet. This matter is being investigated. An interesting point has been raised by Jaeckel,[30] who claims that glass pumps yield better vacua than metal pumps, and suggests catalysis of the decomposition process by the presence of the metal.

It is clear that it is difficult to state the ultimate vacua of diffusion pumps with accuracy. When pumps are required to perform a specific task, the only safe policy is to ascertain first that each particular oil sample is doing what is required of it, and then to take every care of the pump. A control experiment is always necessary where tubulation errors may exist. An increase in published data on every conceivable point associated with the operation of diffusion pumps is necessary so that the scientific aspects of the observed phenomena may become clearer.

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DISCUSSION

Mr. B. D. Power: Mr. Riddiford has complained of unexplained variations in the performances of oil diffusion pumps, though there may be no apparent difference in the arrangement of the pump or test gear, or in the properties of the fluid charge.

We have found that such variations are frequently due to one or more of the pump stages "running wet." Wet running of the top-stage is particularly troublesome. "Wet running" describes the conditions when the bottom edge of the jet cap is covered inside and out with a beading of condensed pumping fluid, and droplets of the fluid fly over as liquid from the jet to the cooled walls. In severe cases external wetting of other parts of the jet assembly may occur. The ill-effects of "wetness" are due to random evaporation from the liquid patches providing a more intense back-streaming flow of vapour than would otherwise be present.

The heat balance in a diffusion pump may be very delicate. In a border-line case slight differences in the way an interior was inserted sufficiently altered the temperature gradient in it to tip the balance between wetness and dryness. The degree of wetness in a wet-running pump can vary from day to day. We have repeatedly found that, when wetness is present, consistent pumping speeds cannot be guaranteed for different pumps of the same design, or for the same pump on different occasions. In one case speeds were reduced by about 70%. Proper measures to ensure dry running of at least the top stage have permitted high speeds to be consistently obtained.

We have less evidence of the influence of wetness upon ultimate pressure, but it is likely that a similar effect exists. High-vacuum baffles depend for their efficiency on the tendency of back-streaming oil molecules from the pump jets to move in long straight paths until they strike a cold baffle plate, and on their complete accommodation to the plate temperature on collision. These two conditions are only approximately fulfilled, so the vapour pressure above the baffle must exceed saturation pressure at the baffle temperature by an amount depending on the quantity and temperature of the back-streaming oil and on the baffle design—and wetness promotes back-streaming.

Author's reply: The figures on ultimate vacua were obtained partly during routine testing of the synchrotron 16 in diffusion pumps, and so were not all made with the same pump. Further, in any event of course the jet assembly must be removed to change an oil charge. However, we have not observed short-term variations in ultimate vacua, and generally a pump, if operated continuously, will perform consistently for many months.

The proposal that the differences in ultimate pressure are due to variations in the nature of the oil samples, and to ageing, is of course a suggestion only, and one would certainly be much happier with an explanation enabling the problem to be more easily dealt with experimentally. Evidence in favour of the proposal is that the rate at which the ionization gauge absorbs the oil decomposition products (quantity S in the table), also varies substantially from sample to sample. Against this it is to be noticed that the effect of an ice trap on the pressure is similar in the two cases of dioctyl sebacate, and also in the two cases of Apiezon oil, which were investigated.

One does not consider that an explanation in terms of change in pumping speed, and temperature of the back-streaming molecules, is sufficient. We, too, have observed variations in speed (of about $\pm 15\%$) between apparently identical pumps, and also for the same pump at different times, and are glad of an explanation for this. This effect is, however, too small to explain the phenomena observed, and certainly does not enter into the discussion if, as suggested by Mr. Power, the ultimate molecules are oil molecules. A more convincing argument than the suggestion of low accommodation coefficients, however, is that the decomposition products of the oil on the top of the wet running jet escape directly into the equipment without having to disentangle themselves from the jet itself, as do decomposition products from the boiler and tower. This argument is supported by the fact that eventually a black "tar" appears on the outside of the jet system.

The problem is obviously one requiring much careful and patient work, and we look forward with interest to the results of Mr. Power's work, when it is complete. The matter is intimately connected with the nature of the ultimate molecules.

Mr. D. R. Goddard: In the interpretation of ultimate pressures as measured by an ionization gauge, has the use of a mass spectrometer to separate the fractions due to partial pressures of oil, water and air been considered? Figures obtained for a 2 in pump using dioctyl sebacate, attached to a mass spectrometer covering a mass range of 0 to 160, show that the residual ion current contains a good proportion of air and water molecules.

Author's reply: In the present case the possibility of significant air- and water-vapour leaks is ruled out by leak-rate considerations. Also, the fact that the ultimate pressure remains constant for months is further evidence against the presence of water-vapour, unless there existed a leak from the water-cooling jacket on the pump. We have not had the time to arrange mass spectrometer equipment, but Mr. Blears has reported elsewhere results of such an investigation. It should be noticed that he observed a mass distribution which was spread over the entire range up to mass 300, which exceeds substantially the range of the measurements quoted.

The estimate of an "average" mass of 160 is not meant to be a precise one, but is supported by the fact that Mr. Blears' distribution is spread widely on both sides of that figure. The sensitivity of an ionization gauge is not exactly the same for all oils, and, further, the gas-scattering effect of the ultimate molecules will depend somewhat on their nature.

Recent advances in vacuum coating plant and techniques

By L. Holland, Research Laboratories W. Edwards and Co. (London), Ltd., London, S.E.26.

Advances in the design of vacuum evaporation plant are shown by reference to a typical plant used for anti-reflection filming of lenses. Methods of removing surface contaminants by ionic and electron bombardment cleaning are reviewed. Recent industrial applications of vacuum coating, including the metallizing of plastic foils and cathode-ray fluorescent screens, are described. '

During recent years an increasing amount of attention has been given to both the properties and uses of thin solid films deposited in a vacuum by the evaporation technique. Fortunately the development of efficient high vacuum equipment designed on engineering lines has kept pace with the interest shown in evaporated films. In this short review it is only possible to deal with some aspects of the advance made in plant construction and give examples of the application of the process by industry.

EVAPORATION PLANT

The difficulties encountered in producing successful coatings with early vacuum plant were mainly due to the low speeds of the available diffusion pumps, but this position is now changed since efficiently baffled high-speed oil diffusion pumps up to 16 in and more in diameter are in normal manufacture. Moreover, the advent of silicone fluids has considerably reduced the risk of oil decomposition by accidental exposure of the diffusion pump, whilst hot, to the atmosphere.
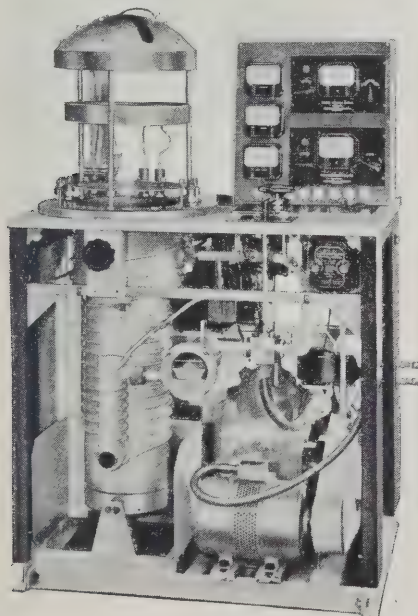
Fig. 1. Interior view of evaporation plant showing the pumping equipment. The chamber apparatus is used for deposition of anti-reflection coatings on glass and includes bombardment electrode rings, radiant heater and lens mounting jig
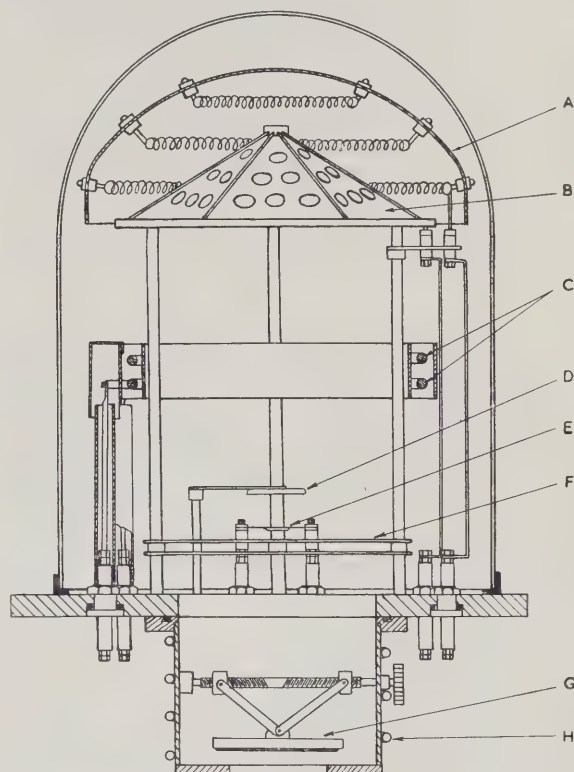
Fig. 2. Chamber-layout for deposition of anti-reflection films on to lenses: *A*, radiant heater; *B*, lens holder; *C*, bombardment rings; *D*, vapour shutter; *E*, evaporation source; *F*, radiation shield; *G*, combined oil baffle and isolation valve; *H*, water cooling coils

Although oil diffusion pumps have largely replaced mercury pumps because they do not require liquid air traps, there is evidence that for some substances the properties of the deposited film are affected by the presence of oil vapour with a pressure of the order of 10^{-6} mm of mercury, e.g. rhodium films deposited on glass in oil systems do not equal in adhesion those produced on mercury plants.

An evaporation plant of a type widely used for depositing anti-reflexion coatings on glass is shown in Fig. 1. The oil diffusion pump is 9 in diameter and has a speed of 1 500 l/sec in the pressure region below 1 μ. The pump is fitted with a booster stage and has a maximum backing pressure of 0·5 mm of mercury so that only a single stage forepump of moderate speed is required for backing the diffusion pump.

The oil baffle *G* (Fig. 2) fitted to the diffusion pump is part of a 12 in diameter high-vacuum valve. When the sealing plate of the valve is raised from its seating it remains parallel to the pump aperture and prevents the direct access of back-streaming oil molecules into the chamber. Oil vapour not impeded by the sealing plate is condensed on to the water-cooled walls of the high vacuum valve. The plant cooling water is fed to the valve before entering the diffusion pump; thus the vapour pressure of oil in the chamber never exceeds the saturated vapour pressure of the oil at the cooling water temperature.

After baffling, the combination has a speed of 1 000 l/sec, which, with a chamber capacity of 100 l, means an extremely high ratio of pumping speed to degassing area. This is of importance for applications such as the deposition of hard anti-reflexion films of magnesium fluoride on to lenses, where the temperature of the lenses and holding fixture is raised to 250°–300° C and the degassing rate thereby increased. Bateson and Bachmeier[1] have drawn attention to the need for fast pumping to produce hard films of magnesium fluoride.

For observation of the plant performance it is valuable to be able to measure the backing pressure, and for this purpose a Pirani gauge is used in the plant described which has the additional advantage that it can be used for leak detection using the hydrogen probe technique. For measuring the chamber pressure an ionization gauge of the Philips cold cathode type is employed since it is sensitive to both gases and vapours and is robust enough for such practical work.

VACUUM SEALS

Continuous maintenance and leak detection was required on earlier plants due to the fragile nature of the waxed seals and greased joints used. Apart from these limitations the presence of such seals in the vacuum chamber was often a nuisance because of their heat sensitivity and the vapours which they liberated. These seals have now been replaced by easily demountable unions and flanges fitted with synthetic rubber "0"-rings.

IONIC BOMBARDMENT CLEANING

To remove surface contaminants remaining on the substrate after chemical cleaning, it is now an established procedure to pass an electrical discharge in the vacuum chamber before deposition is commenced. The glow discharge removes thin contaminating films under bombardment of the ionized gas molecules, and it is generally true to say that, without this preliminary cleaning action, the properties of evaporated coating, i.e. adhesion, reflectivity, etc., are poor. The general clean-up of the vacuum system by the discharge is evident because it is usually accompanied by an initial rise in pressure as degassing is accelerated. It is, therefore, surprising that often very little care is taken to employ effectively this important auxiliary technique. Thus the discharge

should be prevented from (i) penetrating into the diffusion pump oil baffle where the oil can be decomposed, and (ii) bombarding the surfaces of metal holding fixtures which might sputter.

In the coating plant described, the discharge is operated from a high tension transformer, the output of which is connected to the chamber bombardment electrodes via two insulated electrodes in the chamber baseplate (see Fig. 2). The bombarding electrodes, two aluminium rings of equal diameter, are mounted on to a tripod which also carries a lens holder. The bombarding ring diameter is such that it does not obstruct the vapour stream from the evaporation source which is arranged at the centre of the baseplate.

The bombarding rings are separated by a gap which is less than the distance required for sustained ionization, consequently the discharge takes the longer path from the outer surfaces of the rings, which has the effect of concentrating the bombardment in the centre of the chamber, i.e. in the vicinity of the lens holder. Fig. 3

Fig. 3. Glow discharge from ionic bombardment in progress. Two aluminium ring electrodes are used separated from the chamber wall by a shield ring to prevent overheating of the chamber

shows the discharge in operation; the electrode rings are shielded from view by a metal band which stops the discharge from over-heating the chamber wall. The rings are also shielded by an inner earthed band to prevent them from being coated with evaporated metal which might sputter on subsequent cleaning cycles.

In the case of large mirrors careful design of the chamber electrodes is necessary to obtain uniform bombardment, as shown by Benson, Hass and Scott.[2]

ELECTRON BOMBARDMENT

Dunoyer[3] has described a method of removing surface contaminants by electron bombardment. Interes-

in this has been revived in America by Rice[4] and Dimmick,[5] who also used the bombardment for changing the structure of a deposited film. A hot cathode is used for the electron source and the electrons are accelerated towards the substrate surface by making the holding fixture the anode. One method[5] uses a tantalum foil for the cathode, but the author found that this was poisoned badly on a silicone oil diffusion plant in the pressure region 10^{-5} to 10^{-4} mm of mercury. A satisfactory filament can be made by painting thorium oxide on to a tungsten spiral; the thorium oxide is easily renewed should it become poisoned in use. The bombardment can be operated from an alternating supply with the current flowing on each half cycle, but a rectified supply is preferable because it prevents undesirable reverse currents if the pressure in the chamber rises during degassing.

Heating by electron bombardment is a well-known technique and it is a very efficient method of evaporation if the electrons impinge directly on to the surface from which the evaporation is to take place. Smith[6] has recently used this method in a novel way for evaporating thick films of beryllium.

RECENT INDUSTRIAL APPLICATIONS

Recent industrial applications have brought about the development of automatic and continuous coating equipment, and, although these plants are similar in principle to those already described, in some cases the pumping system must be adapted to meet the needs of the application.
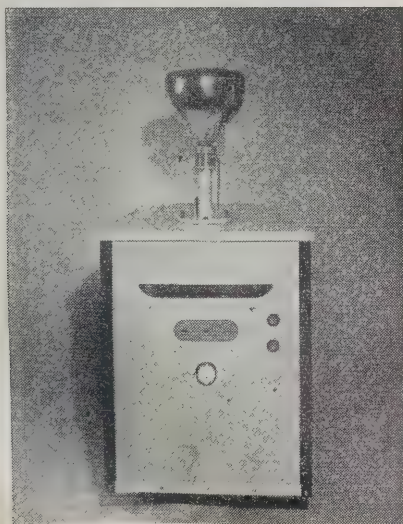
Fig. 4. Automatic coating unit for aluminizing C.R.T. fluorescent screens. The unit contains oil diffusion and rotary pumps with compressed air-operated vacuum valves

Shown in Fig. 4 is an example of an automatic unit used for depositing aluminium on to the fluorescent screens of cathode-ray tubes, a technique which greatly

improves the light output of the tube.[7] The by-pass and high-vacuum valves are compressed-air operated and controlled by a central cam box. The unit can be mounted with others on to a rotary table and the cam box shaft actuated by striker pegs fixed to the floor. Similar units have been made employing magnetic valves..

Another interesting development is the production of large front-surface aluminium mirrors used in the optical systems of television receivers. In the industrial evaporation plant shown in Fig. 5 the mirrors to be coated are supported by frames at each end of the vacuum chamber and, during deposition, are rotated by external motors driving through vacuum shaft seals. Rotation of the mirror greatly simplifies the arrangement of the evaporation sources needed to obtain a uniform deposition. Plants using vacuum locks for the continuous coating of mirrors have also been reported.[8]
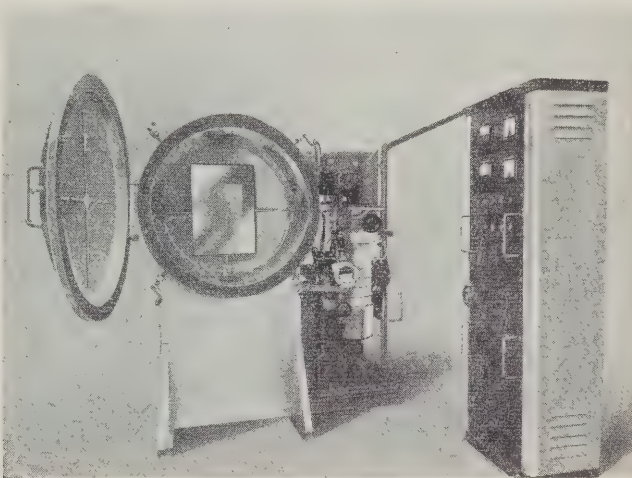
Fig. 5. Large coating plant, 3 ft diameter, used for producing aluminium front surface mirrors. A mirror can be seen in position held in a frame which is rotated during evaporation

Head-lamp reflectors are being produced by depositing aluminium on to a lacquered metal substrate. The metal base of the reflector is finished with a thermo-setting lacquer to produce a specular surface, thereby avoiding expensive metal polishing prior to the deposition of aluminium.

Plastic mouldings are metallized in plants similar to that illustrated in Fig. 5 for decorative and electrical purposes. The plastic mouldings are usually lacquered before coating to improve the surface brilliance and the aluminium protected against wear with either a transparent or tinted lacquer. To obtain an even distribution of the deposited metal on intricate shapes, the mouldings are placed on rotating jigs. Plastic sheeting in roll form can also be metallized by vacuum evaporation, but a special pumping system is needed to remove the volatiles liberated from the foil.[9] The evolution of these

volatiles takes place at a higher pressure than that of normal degassing, in fact in a pressure region where rotary pump speeds are decreasing and normal diffusion pump speeds low. For these reasons the degassing system must be fitted with booster type diffusion pumps (having high speeds at pressures up to 0·5 mm of mercury) and refrigerated traps in the backing line to prevent volatiles from contaminating the rotary pump fluid.

Exposure of the plastic sheet to air after degassing is quite safe as re-absorption of moisture is practically prevented while the material is kept rolled. The pressure obtained during coating is partly determined by the nature of the plasticizer present in the plastic foil; if this is very volatile not only is the vacuum impaired but the plastic properties altered because of evaporation losses.

Zinc and, in some cases, aluminium are being deposited on to condenser paper in vacuum and used to replace the conventional paper and metal foil condenser. The high volatility of zinc makes it possible to deposit conducting films with paper speeds at several hundred ft/min.[10, 11]

A problem with roll coating plants is the design of the evaporation source. On batch coating equipment a resistance-heated tungsten filament, charged with the metal to be volatilized, is invariably used, but this cannot be employed for continuous evaporation due to the limited life of the tungsten filament, which is gradually disintegrated by alloying with the charge, e.g. aluminium and tungsten. Methods evolved to overcome this include automatic feeding of the charge on to the heater so that rapid evaporation takes place, and the use of ceramic crucibles with high frequency heating of the charge.

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DISCUSSION

Mr. A. G. Hayes: The author stated that it is very difficult to evaporate large quantities of aluminium, owing to deterioration of the support. What is the best method that has been found?

Author's reply: As far as I am aware, no completely successful method exists for continuously evaporating large quantities of aluminium. The development of a technique is a most important vacuum coating requirement. Alexander[1] has described a method of feeding aluminium wire on to a tungsten heater, the temperature of the latter being adjusted so that evaporation takes place almost instantaneously. With this arrangement the life of the tungsten heater is extended, but experience shows that the wire feeding rate and the temperature of the heater must be carefully adjusted to prevent spluttering of the molten aluminium.

Although production methods employing high frequency and resistance heated crucibles are being used, certain operational difficulties are experienced. For example, alumina and carbon crucibles react with aluminium in the evaporation temperature region 1 200–1 500° C. The reaction is not always excessive, but the wetting action of aluminium on the surfaces is so high that the walls of the crucible are often fractured by contraction of the aluminium on cooling.

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Melting and sintering of metals in vacuo

By E. D. MALCOLM, B.Sc., A.Inst.P., British American Research Ltd., Wishaw, Lanarkshire

The advantages and applications of melting and sintering in a vacuum are surveyed. The various problems peculiar to the use of a furnace in a vacuum enclosure, and the design of such a furnace suitable for melting 50 to 100 lb of steel are discussed.

In sintering, purifying, heat treating or casting metals some form of atmospheric protection is necessary. Atmospheric attack is practically eliminated in a vacuum, and much of the gas already present in the metal is driven off and may be pumped away.

Vacuum methods have been used in metal purification for over forty years in Germany.[1, 2] The recent advances in industrial high vacuum techniques have given a new impetus to this work and metals of much lower gas content are now economically available. Gas-free metals exhibit increased ductility and toughness. They show high density and freedom from blow holes. Electrical and thermal conductivities are improved.[3, 4]

GAS SOLUTION

A gas may dissolve in a metal in two ways. It may form a simple solution, or it may form a compound with the metal.

For simple solution, Q, the quantity of gas dissolved in a metal, the temperature T, and p the gas pressure over the metal are connected by an equation of the form $Q = kp^{\frac{1}{2}}e^{T}$, where k is an empirical constant.[5] Further at the melting point of the metal there is a sharp rise in solubility.

Sieverts and Krumbhaar[6] have published results on the saturation solubility of hydrogen in various metals. Although equilibrium conditions are never reached in practise Table 1 demonstrates the rapid solution of oxygen and hydrogen in molten nickel.

In the second kind of gas solution oxygen contamination giving the metal oxide is most common. The dissociation pressures p of the oxides and the temperatures of dissociation T are connected by equations of the form $\log p = A - B/T$, where A and B are constants. For iron oxide at $1\,550^\circ$ C the dissociation pressure is 10^{-5} mm, and a temperature of $2\,500^\circ$ C is required to raise this pressure to 1 mm as would be required in a vacuum furnace to drive the reaction effectively towards lower oxygen content.[7, 8] Table 2 gives some comparative figures for vacuum cast copper (carbon deoxydising), and commercial oxygen free copper[9]. Table 3 gives similar results for iron. The analysis for vacuum cast iron is from a 100 lb ingot.[10]

SINTERING

The mechanism of sintering can be explained by a process in which the metals flow viscously under the influence of the opposing forces of surface tension and gas pressure.[11] Three kinds of gas may exist in the pores of a sinter; (a) those which dissolve in the metal

or react with it; (b) those which diffuse through it; (c) those which neither diffuse, dissolve or react with it.

Gases of all three types slow down the sintering process and additionally those of type (a) will give rise to impurities, and of type (c) to porosity.[11, 12]

By sintering in a vacuum enclosure the effects of the gases of all three types can be reduced by a factor of about 10^4. Thus the general result of vacuum sintering is to give a high density compact of improved purity.

Table 1. *Gaseous contamination of molten nickel at various pressures*

(J. H. Moore, N.R.C., unpublished)

Starting material: nickel with gas content:—
oxygen: $1 \times 10^{-3}\%$; nitrogen: $1 \times 10^{-5}\%$;
hydrogen: $3 \times 10^{-5}\%$.

Period of exposure to gas: 10 min.

Oxygen pressure (mm)	Oxygen (%)	Nitrogen pressure (mm)	Nitrogen (%)	Hydrogen pressure (mm)	Hydrogen (%)
0·001	$1\cdot5\times10^{-3}$	27	$1\cdot7\times10^{-4}$	2·2	7×10^{-5}
0·02	4·0	50	3·0	6·7	22
0·10	15·0	760	8·0	33·2	34

Table 2. *Comparative data for vacuum-cast and commercial oxygen-free copper*

Quantity	Commercial oxygen-free copper	Vacuum-cast copper
hydrogen, %	$1\cdot2 \times 10^{-4}$	1×10^{-5}
oxygen, %	$4\cdot5 \times 10^{-4}$	2×10^{-5}
other gases, %	$4\cdot0 \times 10^{-4}$	2×10^{-5}
sulphur, %	$2\cdot3 \times 10^{-3}$	6×10^{-5}
density, gm/cc	8·922	8·930
elect. cond., Int. Units	99·4	100·3
elongation, %	17	20
thermal cond., lb/ft/°F	2 340	2 430

Table 3. *Comparative gas contents for vacuum-cast and electric furnace iron*

Quantity	Electric-furnace	Vacuum-cast
oxygen, %	0·05	0·05
nitrogen, %	0·05	0·001
carbon, %	0·05	0·005
hydrogen, cc/g	0·01–0·03	

VACUUM FURNACES

Turning now to the design of vacuum furnaces, and in particular to a high frequency one (type B of British American Research Ltd.) designed to handle melts of 50 to 100 lb of a non-magnetic steel alloy, the essential requirements are, on the vacuum side: (1) a vacuum tank or enclosure, (2) a pumping system, and on the thermal side: (1) a crucible and heating assembly, (2) a

suitable energy supply, (3) a mould into which to cast the metal.

Fig. 1 shows a sectioned view of the furnace with the exception of the high frequency generator and power and coolant supplies.

VACUUM TANK

The size of the tank is dictated by the amount of energy required to process the melt; 1 000 cm² of tank surface per kilowatt of supply is average. To reduce induction losses stainless steel is used in the fabrication of the tank and all internal fittings. Here the tank has a volume of 50 ft³, and is fitted with three sight glasses, two pumping ports, a power feed through and five
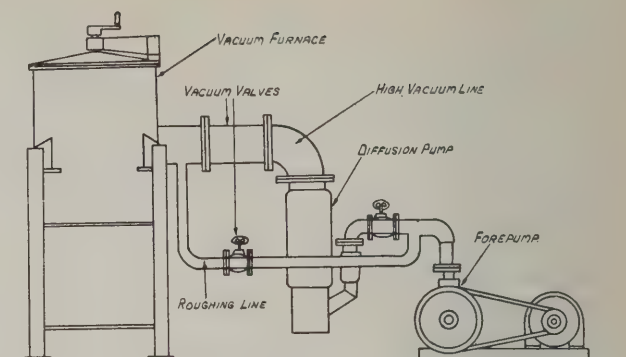
Fig. 2.   Schematic diagram of typical pumping system

Fig. 1.   High vacuum melting furnace

Wilson seals for transmitting motion. The leakage rate is held better than 50 μ/h pressure rise using standard fabrication methods. The tank and tank cover are completely water jacketed.

PUMPING SYSTEM

The main requirement of the pumping system, shown schematically in Fig. 2, is that it have a high pumping capacity in the micron region. In this case it consists of two 6-inch booster type oil diffusion pumps backed by two 100 ft³/min mechanical pumps. The booster pumps have a maximum combined capacity in the

10 to 750 μ range of approximately 55 000 μft³/min (i.e. 2 l/min, air at N.T.P.).

ENERGY SUPPLY

Heating of the charge may be by radiation, conduction or high frequency induction.

Radiation heating is very effective under vacuum and small self-supporting electrical heaters have been used on many occasions. Normal 50 c/s supplies can be used for this type of heater. Retort heating has been used but temperatures are then limited to about 1 100° C by the strength of the retort.

Reche[13] gives a very thorough analysis of induction furnace theory and presents tables from which the crucible and coil constants can be calculated. The crucible containing the metal is surrounded by, and heat insulated from, a water cooled copper coil through which is passed the high frequency current.

The minimum power required for the furnace is that which will cover the surface losses, by radiation and conduction from the crucible and insulation at its maximum temperature. Melting times of about 30 min require approximately 1 kW of generator capacity per pound of steel in the charge.

For melts of under 5 lb, radio frequency generators are generally used, and for larger charges and furnaces motor generators with frequencies in the range 1 000–20 000 c/s are employed. In the type B furnace a 50 kW, 2 000 c/s 400 V generator is used with a twelve turn coil and a 500 μF condenser bank.

High temperature porous insulating refractories have been found suitable as far as outgassing under vacuum is concerned. Mycalex sheet and rod are extensively

used in the coil assembly box as they do not degas and stand fairly high temperatures.

CASTING

Once the metal has been degassed it is undesirable to allow it to solidify in the crucible since (1) cooling times would be very long because of the heat insulation, (2) relatively large pipes are formed and considerable loss of metal results,[14] (3) it is found that a certain amount of residual gas is liberated during the pour, (4) where alloys are being processed segregation may occur.

Fig. 3 shows three methods of bottom pouring. Fig. 3(a) shows perhaps the best method as the metal can fall freely from the crucible. The method shown in Fig. 3(c) requires no mechanical motions, but the material of the fusible plug will contaminate the melt and outgas when heated.
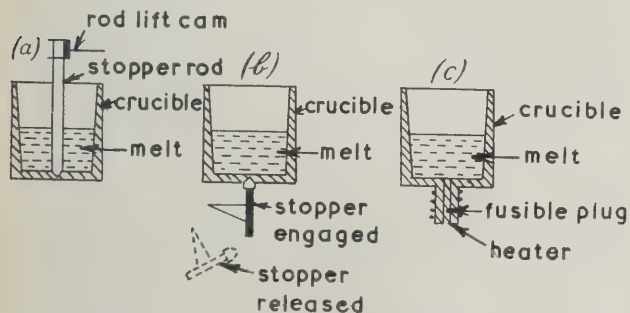
Fig. 3.   Designs for bottom pouring from vacuum melting furnace
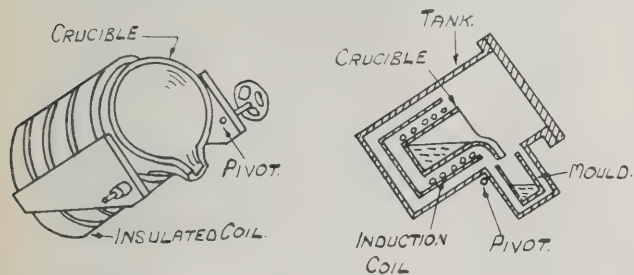
Fig. 4.   Designs for tilt pouring from vacuum melting furnace

Fig. 4 shows two methods of tilt pouring. The first is used where the crucible is mounted in a large vacuum enclosure as in the type B furnace. In the second a great reduction in the volume of the vacuum enclosure has been made and crucible, mould and tank all rotate through 90 degrees. It requires a flexible high-vacuum pumping conduit and to date has been used only on small units (500g maximum) because of this requirement.

In both cases flexible h.f. leads and water-cooling leads are required. This has been achieved by running a multi-stranded braided cable through a rubber or plastic water pipe thus combining the two functions.

MOULDS

Rohn[15] who has built many industrial vacuum melting furnaces reported that sand and other ceramic moulds invariably outgassed badly during pouring, making the maintenance of low pressures difficult and also contaminating the cast. Water-cooled metal moulds are therefore used and copper is superior to steel because of its higher conductivity.

Fig. 5 shows a cross-section of a water-cooled mould for casting 300 lb copper ingots.[9] This mould requires 150–200 gal/min of cooling water during and immediately after pouring. The type B mould for 50 lb of steel (4 in diameter) requires 100 gal/min of cooling water.
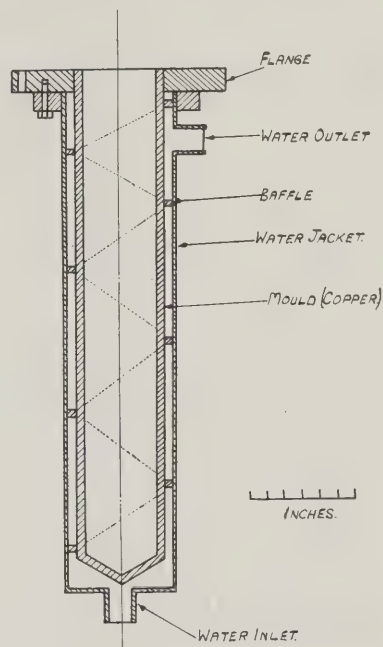
Fig. 5.   Design for water-cooled mould for use in vacuum casting

To allow the gas which is liberated during pouring to be pumped off the casting is done slowly (about 3 min for 300 lb copper). Slow pouring also prevents the formation of long pipes.[9]

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Freeze drying—vacuum sublimation

By L. G. BECKETT, Research Laboratories, W. Edwards & Co. (London) Ltd., London, S.E.26.

A short account is given of the principles involved in the dehydration of heat sensitive materials by sublimation from the frozen state. References are made to drying rates and important design features of commercial apparatus.

The process of freeze drying is used for low temperature desiccation of heat sensitive biological materials not readily dried by other processes, so that they may be preserved without spoilage. Typical of the materials dried by this process are blood plasma, vaccines, living bacterial and virus suspensions, antibiotics, tissues and various food stuffs. The process involves preliminary freezing with subsequent sublimation drying.

FREEZING

It is desirable that freezing should be as rapid as possible, slow freezing invariably leads to partial separation and hence concentration with attendant spoilage of the product. For short drying cycles it is necessary that the material should have the maximum possible exposed surface and that this be of minimum thickness; this is conveniently obtained by freezing to form a hollow cylinder. Two basic freezing techniques are used: (*a*) freezing by some form of external refrigeration, and (*b*) freezing by rapid evaporation of solute under reduced pressure. Various methods of freezing by external refrigeration are practised,[1,2] but in each case they necessitate apparatus additional to the drying equipment (Fig. 1). Freezing under vacuum by self-evaporation is an attractive alternative if it were not for the difficulty of foaming. This can, to some extent, be controlled by gradual reduction of pressure but, even with care, liquids degas violently prior to freezing, with dispersal of the material which if toxic is a serious hazard to the operators.

Greaves introduced a method of suppressing frothing by centrifugal force.[3] In this technique, the liquid containers are mounted in a carrier (Fig. 2) which is rotated at moderate speed within the drying chamber before and during the early stages of exhaustion, until the material is frozen. Rapid freezing occurs without foaming with the additional advantage of freezing the material into thin shells or wedges for high drying rates. Approximately 20% of the water evaporates before freezing occurs, but fortunately, the evaporation rate is so rapid that "spoilage" is insignificant. The efficient trapping of the large volumes of water vapour produced so quickly necessitates very high rates of heat extraction

from a refrigerated condensing surface or high absorbing capacity (and heat dissipation) by chemical or physical absorbants.

Throughout drying the solution is maintained frozen by the heat required for sublimation, approximately 680 cal/g (2845 J/g). This represents a very high refrigerating effect, to counteract which without melting, provides a difficult heating problem.

VACUUM PUMPING SYSTEMS

The vacuum pumping systems for freeze drying must provide for: (*a*) the initial rapid exhaustion from atmospheric pressure to operational pressures (normally 2·5–0·05 mm of mercury) and thereafter for the removal
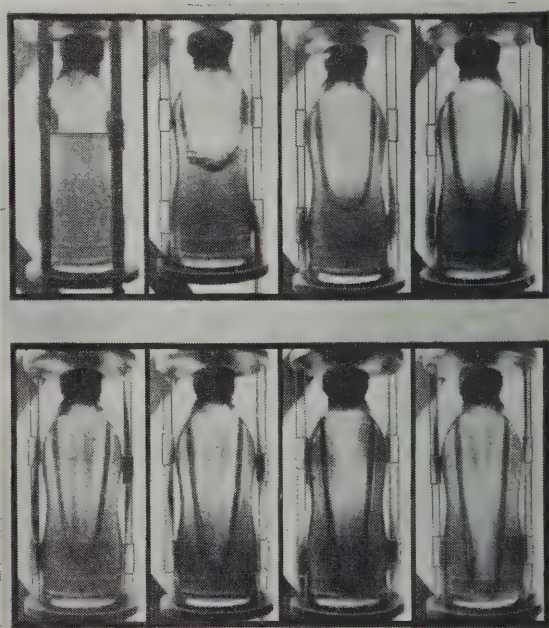
Fig. 1. Vertical spin freezing. Stages in vortex formation during the spinning operation of blood plasma prior to freezing. The material freezes in the shape shown in the bottom right-hand illustration

of traces of permanent gases released from solution during the drying process; and (*b*) the removal of large volumes of water vapour.
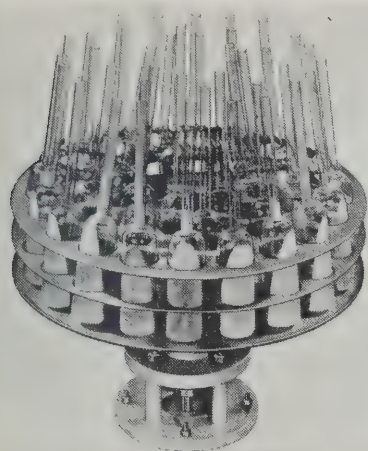
Fig. 2.
Centrifuge assembly with ampoules containing frozen material. Note the wedge formation of the ice

Vacuum pumps are available which will satisfy both these requirements; the commonest for large processing units is the steam ejector, but the cost of the steam is often a prohibitive factor. Mechanical oil-sealed rotary pumps without traps have been used for freeze drying, the water entrained in the oil of these pumps being removed by centrifugal action or by thermal stripping. Except for micro-work, mechanical pumps alone are uneconomic owing to their relatively small pumping speeds at low pressures.

Modern freeze drying equipment utilizes a mechanical pump for the initial phase and in combination with either a chemical desiccant or refrigerated condensing surface for the second drying phase.

ABSORPTION BY CHEMICAL DESICCANTS

Chemical desiccants are well suited for laboratory equipment because of simplicity and low initial cost of the plant. The choice of desiccant requires careful consideration, not necessarily for performance but for operational convenience. Plant design is greatly influenced by the desiccating value of the desiccant in a vacuum. The smaller this is the larger must be the vacuum chamber to hold it with corresponding increase in the size of the pump. The two commonest desiccants used for freeze drying are anhydrous calcium sulphate and phosphorus pentoxide. For satisfactory vacuum dehydration a minimum of 90 g of the former and 3 g of the latter are required per gram of water removed. Many other desiccants have been proposed and used. Probably the most interesting is a solution of lithium chloride which, after cooling to a temperature just below 0° C is sprayed into the vacuum system, where it entrains and absorbs water vapour. The water-enriched solution is then removed from the vacuum system and thermally stripped. After cooling, the solution is re-cycled through the vacuum chamber.

REFRIGERATED CONDENSERS

Because of its high vapour pumping speed and relatively high thermal efficiency, the refrigerated condenser is the system most widely used in commercial apparatus for eliminating water from the drying system. High water vapour pumping speeds are essential as the specific volume of water vapour at 0·1 mm of mercury is approximately 10 000 l/g. An installation designed for drying 2 l of aqueous material per hour at a pressure of 0·1 mm of mercury must, therefore, be provided with a water vapour pumping speed of 5 000 l/sec. In many batch type processes a static condenser system is used (Fig. 3), this requires short shut down periods for deicing. Continuous type condensers have been constructed in the form of a hollow refrigerated cylinder, the inner surface of which is exposed to the vacuum system. Ice condensed on this surface is continually
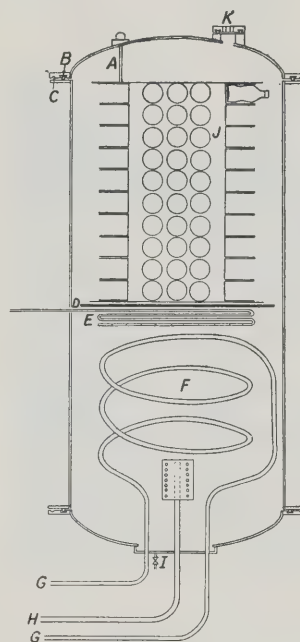
Fig. 3.
A. Thermostat control.
B. Rubber sealing ring.
C. Vacuum proving groove (using tracer gas).
D. Radiation shield.
E. Defrosting coil.
F. Water vapour condenser.
G. Refrigerated brine pipes.
H. Vacuum pump connection.
I. Water drain valve.
J. Electrically heated bottle header.
K. Glass to metal seals for electrical supply

removed by rotating scraping vanes and discharged to atmosphere via a vacuum tight lock (Fig. 4).

Factors influencing the design of freeze drying systems utilizing refrigerated condensers are: (*a*) pressure and temperature difference between product and condenser surface; (*b*) the saturation vapour pressure of ice at the condensing surface temperature, and the ultimate pressure obtainable with the vacuum pump employed; (*c*) thermal efficiency, simplicity and reliability of refrigerating system.

The conditions for (*a*) and (*b*) are determined by the maximum temperature to which the product can be safely raised. For the bulk of biological materials little is gained by employing condenser surface temperatures below − 40° C. The thermal efficiency and reliability of a mechanical refrigerating system increases as the evaporator (cold surface) temperature is raised and as it approaches the temperature of the condensing system.

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Furthermore, temperatures of −40° C are readily obtained with simple refrigerating machines.

A further consideration for condition (*b*) is the matching of pump vacuum performance with saturation vapour pressure of ice at the temperature of the condenser surface. At −40° C ice has a vapour pressure of 0·09 mm of mercury which for practical purposes is equal to the ultimate pressure obtainable with a single stage rotary pump. If, therefore, a drying system has a condenser operated at −40° C and is provided with pumping equipment which will produce pressures much lower than 0·09 mm of mercury, ice will be resublimed from the condenser and pass into the pumps, contrary, of course, to requirements.

On a laboratory scale a mixture of solid carbon dioxide and alcohol forms a convenient method of refrigerating a water vapour condensing surface. In a typical glass laboratory freeze dryer approximately 6 g of solid carbon dioxide are required for each gram of ice condensed.
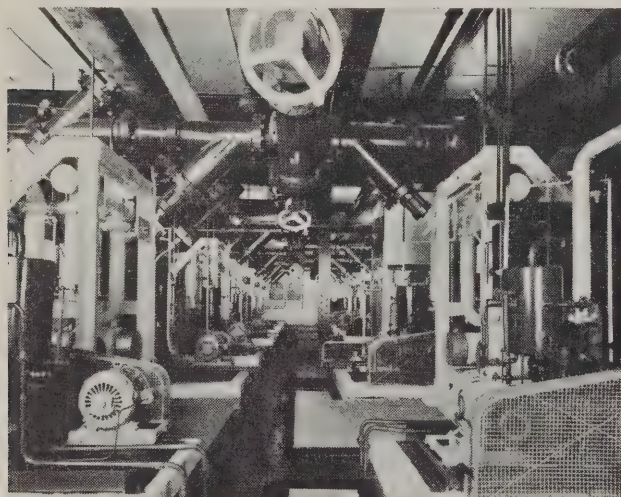
Fig. 4. Pump installation for penicillin plant. The pipe lines connect to scapping vane refrigerated condensers shown to the extreme right and left of the illustration. In front of these are mounted single stage vacuum pumps

DRYING RATES AND STAGES

It is not yet possible mathematically to predict drying rates for biological material. For estimation, however, it can be assumed that the ice surface of a frozen product exposed to vacuum will recede at 0·2 mm to 1 mm/per hour.[2] Under similar drying conditions, human blood plasma (having a solids content of about 10%), to reach a moisture content of approximately 1%, requires 4–5 times the period necessary for the complete sublimation of a similar volume of ice.

The drying rate of a frozen protein solution is substantially linear down to a moisture content of about 5%, when ice visible to the unaided eye will have normally disappeared from the product. Thereafter, the drying rate proceeds exponentially. To reduce the residual moisture content of the dried product from 5% to 0·5%

may require up to 20% or more of the time required to reduce the moisture content from 100% to 5%.

Owing to the decrease in drying rate near complete dehydration it is usual to employ two drying systems, a primary drying system using a refrigerated water vapour condenser and in which the moisture content is reduced to approximately 1%, and a secondary drying system using a chemical desiccant, in which the moisture content is reduced to the required low value, normally about 0·5%. This division of the drying cycle ensures optimum loading and output for a primary drying installation of given size and is of considerable economic importance.

CONCLUSION

Freeze drying is the ideal method for the preservation of labile medical and veterinary preparations by dehydration as it results in the minimum amount of damage to the product, or to use the current biological term, "minimal spoilage."

The great increase in the application of this technique within recent years has removed it from a laboratory to an industrial scale. The economic design of plant therefore becomes most dependent on close co-operation between biologist, physicist, and engineer, if their often conflicting requirements are to be satisfied.

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DISCUSSION

Dr. M. Pirani: Mr. Beckett stated that some vacuum engineers recommend pumping systems for freeze drying which, in his opinion, incorporate an unnecessary feature. As he pointed out, with the "usual" trap temperature of −40° C, the ice crust on its surface has a vapour pressure of 0·1 mm. Therefore the vapour stream pump serves only to transport water vapour into the mechanical pump. It does not cause an improved drying rate. However, in certain large industrial freeze drying plants the trap is operated at a temperature between −70° and −100° C, so that the water vapour pressure is less than 2 μ. Under these conditions evaporation from the condenser is negligible.

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